

Functional Approach to Electrodynamics of Media

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Abstract

In this article, we put forward a new approach to electrodynamics of materials. Based on the identification of induced electromagnetic fields as the microscopic counterparts of polarization and magnetization, we systematically employ the mutual functional dependencies of induced, external and total field quantities. This allows for a unified, relativistic description of the electromagnetic response without assuming the material to be composed of electric or magnetic dipoles. Using this approach, we derive universal (material-independent) relations between electromagnetic response functions such as the dielectric tensor, the magnetic susceptibility and the microscopic conductivity tensor. Our formulae can be reduced to well-known identities in special cases, but more generally include the effects of inhomogeneity, anisotropy, magnetoelectric coupling and relativistic retardation. If combined with the Kubo formalism, they would also lend themselves to the ab initio calculation of all linear electromagnetic response functions.

Keywords: Polarization, Magnetization, First-principles calculation, Bi-anisotropy, Multiferroics, Relativity

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1. Introduction

Standard approach and its limitations. Electrodynamics of media aims at describing the response of a material probe to external electromagnetic perturbations [1–4]. For this purpose, Maxwell’s equations alone are not enough, because these relate only the electromagnetic fields to their sources, but do not describe the reaction of currents and charges in the material to the electromagnetic fields. Therefore, the usual approach to electrodynamics of media is to introduce additional field quantities besides the electric field \mathbf{E} and the magnetic induction \mathbf{B} . These additional fields are the polarization \mathbf{P} and the magnetization \mathbf{M} , as well as the displacement field \mathbf{D} and the magnetic field \mathbf{H} . They are interrelated by

$$\mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P}, \quad (1.1)$$

$$\mathbf{H} = \frac{1}{\mu_0} \mathbf{B} - \mathbf{M}, \quad (1.2)$$

where ε_0 and μ_0 denote the vacuum permittivity and permeability respectively. The fields \mathbf{P} and \mathbf{M} are interpreted as electric and magnetic dipole densities inside the material. The so-called *macroscopic Maxwell equations* are then written in the following form:

$$\nabla \cdot \mathbf{D} = \rho_{\text{ext}}, \quad (1.3)$$

$$\nabla \times \mathbf{H} - \frac{\partial \mathbf{D}}{\partial t} = \mathbf{j}_{\text{ext}}, \quad (1.4)$$

$$\nabla \cdot \mathbf{B} = 0, \quad (1.5)$$

$$\nabla \times \mathbf{E} + \frac{\partial \mathbf{B}}{\partial t} = 0, \quad (1.6)$$

where ρ_{ext} and \mathbf{j}_{ext} are the external charge and current densities. Traditionally, these were identified with the free sources as opposed to those bound in the medium [3, 4]. In modern treatments, however, the sources are split into external and induced contributions [5–11]; see in particular the remarks in Ref. [1, pp. 261 f.]. The general equations (1.3)–(1.6) are complemented by material-specific *constitutive relations*, which in the simplest case read

$$\mathbf{P} = \varepsilon_0 \chi_e \mathbf{E}, \quad (1.7)$$

$$\mathbf{M} = \chi_m \mathbf{H}, \quad (1.8)$$

with electric and magnetic susceptibilities χ_e and χ_m . Taken together, the equations (1.1)–(1.8) can be used to determine the electric and magnetic fields inside the material medium in terms of the external sources. They are usually supposed to hold on a macroscopic scale, i.e., \mathbf{E} and \mathbf{B} represent suitable averages of the microscopic fields, while \mathbf{P} and \mathbf{M} are regarded as average dipole moments of macroscopic volume elements [1–4, 12]. We will henceforth refer to this approach as the *standard approach* to electrodynamics of media. Traditional textbooks (such as [3, 4]) set up this approach and derive Eqs. (1.3)–(1.6) based on the assumption of microscopic dipoles induced by the electromagnetic fields, as for example in the Clausius–Mossotti model [13]. Such simplified models have been ubiquitous in the description of the electromagnetic response of solids for more than a century since Maxwell’s original work in the eighteen sixties [14, 15].

However, two modern developments in solid-state physics have put severe limitations on the standard approach: the quest for an *ab initio* description of material properties [6, 16–18] and the discovery of new materials with exotic electromagnetic responses. As for the first point, the Modern Theory of Polarization [19–21] has raised conceptual questions about the interpretation of \mathbf{P} and \mathbf{M} as electric and magnetic dipole densities. It has been shown that the localized polarizable units of the Clausius–Mossotti model are in stark contrast to the actual delocalized electronic charge distributions of real materials, and hence this model fails in most cases to describe polarization effects adequately [20]. More fundamentally, it was argued that the polarization of a crystalline solid cannot, even in principle, be defined as a bulk quantity in terms of periodic electronic charge distributions, and instead the polarization *change* in a typical measurement setup was defined in terms of a macroscopic charge flow in the interior of the sample [19, 20]. Regarding the second point, with the advent of metamaterials [22–24] and in particular bi-anisotropic media with magnetoelectric coupling [25–27], it became increasingly important to generalize the constitutive relations (1.7)–(1.8) and to reformulate the theory of electromagnetic responses in a more systematic way [28, 29]. Since often the properties of these materials are determined by nanoscale resonant inclusions (“meta-atoms”) [30], macroscopic averaging procedures have also come under intense investigation [31–33]. Further systems with magnetoelectric coupling include the prototype material Cr_2O_3 [34–36], single-phase multiferroics [37–40], composite multiferroics and multiferroic interfaces [41–44] as well as systems with Rashba spin-orbit coupling [45–47]. Topological insulators show the topological magnetoelectric effect

[48–50], which becomes observable if the surface response is separated from the bulk contribution by breaking the time-reversal symmetry [51, 52], for example, through the introduction of ferromagnetism [53–55].

Functional Approach. To overcome the limitations of the standard approach, we will develop in this paper the Functional Approach to electrodynamics of media. This approach identifies induced electromagnetic fields as the microscopic counterparts of macroscopic polarizations and interprets them as functionals of the external perturbations. Thus, we start from the following identifications *on a microscopic scale*:¹

$$\mathbf{P}(\mathbf{x}, t) = -\varepsilon_0 \mathbf{E}_{\text{ind}}(\mathbf{x}, t), \quad (1.9)$$

$$\mathbf{D}(\mathbf{x}, t) = \varepsilon_0 \mathbf{E}_{\text{ext}}(\mathbf{x}, t), \quad (1.10)$$

$$\mathbf{E}(\mathbf{x}, t) = \mathbf{E}_{\text{tot}}(\mathbf{x}, t), \quad (1.11)$$

and

$$\mathbf{M}(\mathbf{x}, t) = \mathbf{B}_{\text{ind}}(\mathbf{x}, t)/\mu_0, \quad (1.12)$$

$$\mathbf{H}(\mathbf{x}, t) = \mathbf{B}_{\text{ext}}(\mathbf{x}, t)/\mu_0, \quad (1.13)$$

$$\mathbf{B}(\mathbf{x}, t) = \mathbf{B}_{\text{tot}}(\mathbf{x}, t). \quad (1.14)$$

¹The analogy between $\mathbf{P}(\mathbf{x})$, $\mathbf{M}(\mathbf{x})$ and electromagnetic fields generated by internal sources has been called “a deceptive parallel” in Ref. [4, Section 4.3.2, 6.3.2], because it implies in particular that $\nabla \cdot \mathbf{M}(\mathbf{x}) = 0$. The bar magnet with uniform magnetization parallel to its axis is considered an example where this condition does not hold. There, the magnetization is assumed to be constant inside a finite cylinder and zero outside, and hence a nonvanishing divergence appears at the top and bottom surfaces. However, the so defined field $\mathbf{M}(\mathbf{x})$ is not observable itself and indeed only serves to determine by $\nabla \times \mathbf{M}(\mathbf{x}) = \mathbf{j}(\mathbf{x})$ the macroscopic current, which is localized at the lateral surface of the cylinder. Any curl-free vector field can be added to $\mathbf{M}(\mathbf{x})$ without changing this information, fact which has been referred to as “gauge freedom” of the magnetization in Refs. [56–58]. A careful examination of the procedure described in Ref. [4] shows that the real, observable magnetic field is given in terms of the current by $\nabla \times \mathbf{B}(\mathbf{x}) = \mu_0 \mathbf{j}(\mathbf{x})$ and $\nabla \cdot \mathbf{B}(\mathbf{x}) = 0$, or in terms of the uniform magnetization by the transverse part $\mathbf{B}(\mathbf{x}) = \mu_0 \mathbf{M}_{\text{T}}(\mathbf{x})$. Similarly, in electrostatics the observable electric field is given in terms of the uniform polarization of a macroscopic body by the longitudinal part $\mathbf{E}(\mathbf{x}) = -\mathbf{P}_{\text{L}}(\mathbf{x})/\varepsilon_0$. Such problems actually do not concern electrodynamics of media but the determination of electromagnetic fields as generated by macroscopic surface currents or charges. Electrodynamics of media concerns the question of how these surface charges or currents can be induced by external fields.

These identifications are common practice in semiconductor physics (cf. [11, p. 33, footnote 14]) and in electronic structure physics (cf. [59, Appendix A.2]). The quantities on the right hand side of Eqs. (1.9)–(1.14) refer to the induced, the external and the total microscopic fields, respectively (see Section 5 and cf. [1, 56]). By *microscopic* fields we mean that these are derived from microscopic charge and current distributions, which in turn are derived from continuous quantum (many-body) wave functions. Explicitly, the induced fields are related to the induced charges and currents by the microscopic Maxwell equations

$$-\nabla \cdot \mathbf{P}(\mathbf{x}, t) = \rho_{\text{ind}}(\mathbf{x}, t), \quad (1.15)$$

$$\nabla \times \mathbf{M}(\mathbf{x}, t) + \frac{\partial}{\partial t} \mathbf{P}(\mathbf{x}, t) = \mathbf{j}_{\text{ind}}(\mathbf{x}, t), \quad (1.16)$$

$$\nabla \cdot \mathbf{M}(\mathbf{x}, t) = 0, \quad (1.17)$$

$$-\nabla \times \mathbf{P}(\mathbf{x}, t) + \frac{1}{c^2} \frac{\partial}{\partial t} \mathbf{M}(\mathbf{x}, t) = 0. \quad (1.18)$$

Analogously, the external fields are related to the external sources by the following equations (cf. [5, p. 4]):

$$\nabla \cdot \mathbf{D}(\mathbf{x}, t) = \rho_{\text{ext}}(\mathbf{x}, t), \quad (1.19)$$

$$\nabla \times \mathbf{H}(\mathbf{x}, t) - \frac{\partial}{\partial t} \mathbf{D}(\mathbf{x}, t) = \mathbf{j}_{\text{ext}}(\mathbf{x}, t), \quad (1.20)$$

$$\nabla \cdot \mathbf{H}(\mathbf{x}, t) = 0, \quad (1.21)$$

$$\nabla \times \mathbf{D}(\mathbf{x}, t) + \frac{1}{c^2} \frac{\partial}{\partial t} \mathbf{H}(\mathbf{x}, t) = 0. \quad (1.22)$$

External and induced sources are not necessarily located “outside” or “inside” the material probe respectively. Instead, this distinction implies that the induced fields are generated by the degrees of freedom of the medium, whereas the external fields are associated with degrees of freedom which do not belong to the medium. In particular, this applies to external sources which can be controlled experimentally [56]. Thus, we use the term *external fields* in precisely the same sense as in classical or quantum mechanics, where one considers particles moving in external fields as opposed to particles moving in the fields generated by themselves.

In view of the above identifications, Eqs. (1.1)–(1.2) hold as exact identities in our approach simply by the definition of the total fields as the sum of the external and the induced fields. (This is in contrast to other approaches such as the multipole theory [60], where Eqs. (1.1)–(1.2) are regarded as first order approximations.) Moreover, Eqs. (1.3)–(1.4) are none other than the inhomogeneous Maxwell equations relating the external fields to the external sources, while Eqs. (1.5)–(1.6) are the homogeneous Maxwell equations for the total fields (cf. [11, Eqs. (2.124)]). In particular, these equations hold in general on a microscopic scale, and they do not need to be derived as in the standard approach (based on induced electric and magnetic dipoles, cf. [3, 4]). Macroscopically averaged fields defined by

$$\langle \mathbf{E} \rangle(\mathbf{x}, t) = \int d^3\mathbf{x}' f(\mathbf{x} - \mathbf{x}') \mathbf{E}(\mathbf{x}', t) \quad (1.23)$$

(with $f(\mathbf{x})$ being smooth, localized at $\mathbf{x} = 0$ and ranging over distances large compared to atomic dimensions in the material) satisfy again Maxwell equations with the averaged sources, because the averaging procedure commutes with the partial derivatives [12]. In particular, this implies that any fundamental relation between the microscopic electromagnetic fields which is derived from the Maxwell equations will hold for the macroscopically averaged fields as well, a conclusion which applies especially to the universal response relations derived in Section 6.

Our main focus in this article is on the microscopic electromagnetic response functions, which are represented by functional derivatives of induced electric and magnetic fields with respect to external perturbations. It is well-known that these are not independent of each other, and hence it is possible to formulate electrodynamics of media as a *single susceptibility theory* [29], where all linear electromagnetic response functions are derived from a single response tensor. For example, in magneto-optics it has been suggested to introduce an effective permittivity tensor relating the \mathbf{D} and \mathbf{E} fields while setting $\mathbf{H} = \mathbf{B}/\mu_0$ identically [61–63]. By contrast, here we stick to the usual definition of electromagnetic response functions and relate each of them to the microscopic conductivity tensor, a possibility which has already been mentioned in Ref. [5] (see also [56, 64]). Starting from the model-independent definitions (1.9)–(1.14), we will thereby obtain *universal response relations*, which can be used to study the electromagnetic response not only of solids, but also liquids, single atoms or molecules, or even the vacuum of quantum electrodynamics (see Section 5.2).

Comparison to the literature. Our main motivation for the identifications (1.9)–(1.14) comes from microscopic condensed matter physics in general and ab initio electronic structure theory in particular [6–10, 65]. There, it is common practice to introduce a microscopic dielectric function (or permittivity) $\varepsilon_r = \varepsilon_r(\mathbf{x}, t; \mathbf{x}', t')$ by means of the defining equation

$$\varphi_{\text{tot}} = \varepsilon_r^{-1} \varphi_{\text{ext}} . \quad (1.24)$$

Here, φ_{tot} and φ_{ext} denote the total and external scalar potentials respectively, and the products refer to non-local integrations in space and time. The total potential is in turn defined as the sum

$$\varphi_{\text{tot}} = \varphi_{\text{ext}} + \varphi_{\text{ind}} . \quad (1.25)$$

It has been noted already (see, e.g., [9, Section 6.4] and [66, Section 4.3.2]) that these definitions are *analogous* to electrodynamics of media provided that one uses the identifications (1.10) and (1.11). The Functional Approach to electrodynamics of media promotes these analogies to *definitions* of the microscopic fields $\mathbf{D}(\mathbf{x}, t)$ and $\mathbf{E}(\mathbf{x}, t)$, and generalizes them via the further identifications (1.9) and (1.12)–(1.14). Consequently, the microscopic dielectric tensor will be defined by (see Section 6)

$$\mathbf{E}_{\text{tot}} = (\overset{\leftrightarrow}{\varepsilon}_r)^{-1} \mathbf{E}_{\text{ext}} , \quad (1.26)$$

which obviously generalizes (1.24). We further note that in the textbook of Fließbach [1], Eqs. (1.9)–(1.14) are deduced for the macroscopically averaged fields in the special case of homogeneous, isotropic media, while in our approach these equations represent the most general definition of microscopic (electric and magnetic) polarizations.

Microscopic approaches to electrodynamics of media have already been described by Hirst [57] and in the so-called premetric approach by Truesdell and Toupin [67] as well as Hehl and Obukhov [68, 69] (see also the textbook by Kovetz [70]). While we have in common the microscopic interpretation of Maxwell’s equations in media, the Functional Approach differs from the above approaches in other respects: (i) By distinguishing between induced and external (instead of bound and free) charges and currents, it is independent of any assumption about the medium. (ii) While the premetric approach distinguishes between “excitations” (\mathbf{D} and \mathbf{H}) and “field strengths” (\mathbf{E} and \mathbf{B}) [71], we only consider electromagnetic fields produced

by different (induced, external or total) sources. Besides the electromagnetic fields, there are no further fields in our approach, which would be of a physically or mathematically different nature, or which would transform differently under coordinate changes. (iii) We provide by Eqs. (1.9) and (1.12) a *unique* definition of the microscopic polarization and magnetization without referring to the constitutive relations of the material. Therefore, the Functional Approach is also suitable for deriving universal response relations, as we will show in Section 6. (iv) While in the premetric approach the linear electromagnetic response is described by a fourth-rank constitutive tensor with 36 components [68, 72], we rely on the functional dependence of the induced current on the external vector potential (see Section 5.1). In fact, we will show that the 9 spatial components of the current response tensor are sufficient to describe the linear response of any material. Moreover, we will derive in Section 5.4 a closed expression for the constitutive tensor in terms of the current response tensor.

Finally, we note that our approach is consistent with the Modern Theory of Polarization [19–21]. This approach relies on a fundamental equation for the *change* in macroscopic polarization in terms of a transient current through the sample given by (cf. also [6, Eq. (22.4)] and [73–75])

$$\mathbf{P}(\Delta t) - \mathbf{P}(0) = \int_0^{\Delta t} dt \mathbf{j}_{\text{ind}}(t), \quad (1.27)$$

where

$$\mathbf{j}_{\text{ind}}(t) = \frac{1}{V} \int d^3\mathbf{x} \mathbf{j}_{\text{ind}}(\mathbf{x}, t), \quad (1.28)$$

and V denotes the volume of the sample. Our identification of induced electromagnetic fields with electric and magnetic polarizations is indeed consistent with this approach, because Eq. (1.27) can be derived classically from the Maxwell equations (1.15)–(1.18) for the microscopic induced fields: First, as the Modern Theory of Polarization focuses on the change of polarization due to *charge transport*, we may restrict attention to the longitudinal part of the current. Differentiating Gauss’ law for the induced electric field with respect to time and using the continuity equation yields

$$-\nabla \cdot \frac{\partial}{\partial t} \mathbf{P}(\mathbf{x}, t) = \frac{\partial}{\partial t} \rho_{\text{ind}}(\mathbf{x}, t) = -\nabla \cdot (\mathbf{j}_{\text{ind}})_{\text{L}}(\mathbf{x}, t). \quad (1.29)$$

This shows directly the identity of the longitudinal parts

$$\frac{\partial}{\partial t} \mathbf{P}_{\text{L}}(\mathbf{x}, t) = (\mathbf{j}_{\text{ind}})_{\text{L}}(\mathbf{x}, t), \quad (1.30)$$

from which one concludes Eq. (1.27) by spatial integration over the sample volume. The importance of the formula (1.27) lies in the fact that it can be reexpressed—assuming a periodic current distribution in a crystalline solid—through the current of a single unit cell [20],

$$\mathbf{j}_{\text{ind}}(t) = \frac{1}{V_{\text{cell}}} \int_{\text{cell}} d^3\mathbf{x} \mathbf{j}_{\text{ind}}(\mathbf{x}, t). \quad (1.31)$$

Hence, the polarization change can be defined as a bulk quantity, which in turn can be inferred from the knowledge of lattice-periodic Bloch wave functions. Therefore, this quantity is also accessible from the results of modern ab initio computer simulations [76–78].

Organization of the paper. We start in Section 2 by reviewing some aspects of classical electrodynamics which are necessary for this paper.

In Section 3, we analyze the initial value problem for both the scalar wave equation and the Maxwell equations. Subsequently, we construct the most general form of the tensorial electromagnetic Green function and discuss its special forms which correspond to special gauge conditions.

Using the tensorial Green function, we express in Section 4 the electromagnetic vector potential as a functional of the electric and magnetic fields. By the help of this *canonical functional*, we then define the notion of *total functional derivatives* with respect to electric or magnetic fields. Sections 3–4 are technical in nature, but the resulting formulae for the vector potential are of central importance for our formulation of electrodynamics of media.

In Section 5, we introduce the general electromagnetic response formalism based on the functional dependence of the induced four-current on the external four-potential. We propose alternative response functionals and thereby establish the connection to the Schwinger–Dyson equations in quantum electrodynamics and the Hedin equations in electronic structure theory. Furthermore, we derive a closed expression for the fourth rank constitutive tensor used in the premetric approach by Hehl and Obukhov [68, 72] in terms of the fundamental response functions.

In Section 6, we first argue that physical response functions have to be identified with total functional derivatives. Based on this, we derive universal (i.e. model- and material-independent) analytical expressions for the microscopic dielectric tensor, the magnetic susceptibility and the magnetoelectric coupling coefficients in terms of the optical conductivity. We further discuss

the intricate problem of expanding the induced electric and magnetic fields in terms of the external fields.

In the closing Section 7, we investigate the various empirical limiting cases of our universal response relations. In particular, we show that these reduce to well-known identities in a non-relativistic limit and in the special case of homogeneous, isotropic materials.

2. Classical electrodynamics

2.1. Notations and conventions

Metric tensor. For the Minkowski metric we choose

$$\eta_{\mu\nu} = \eta^{\mu\nu} = \text{diag}(-1, 1, 1, 1). \quad (2.1)$$

This convention is particularly useful in condensed matter physics, as one can directly read out a spatial three-tensor χ_{ij} from a Minkowski four-tensor $\chi^\mu{}_\nu$ without distinguishing between covariant and contravariant indices (i.e. $\chi_{ij} = \chi^i{}_j$). Consequently, all spatial tensors will be written with covariant (lower) indices. Furthermore, we adopt the convention of summing over all doubly appearing indices (even if both are lower case).

Fourier transformation. With $x^\mu = (ct, \mathbf{x})^\text{T}$ and $k_\mu = \eta_{\mu\nu}k^\nu = (-\omega/c, \mathbf{k})$, we have

$$kx \equiv k_\mu x^\mu = -\omega t + \mathbf{k} \cdot \mathbf{x}. \quad (2.2)$$

We define the Fourier transform of a field quantity $\rho(x) = \rho(\mathbf{x}, t)$ and its inverse as

$$\rho(\mathbf{k}, \omega) = c \int \frac{d^3\mathbf{x}}{(2\pi)^{3/2}} \int \frac{dt}{(2\pi)^{1/2}} \rho(\mathbf{x}, t) e^{i\omega t - i\mathbf{k} \cdot \mathbf{x}}, \quad (2.3)$$

$$\rho(\mathbf{x}, t) = \frac{1}{c} \int \frac{d^3\mathbf{k}}{(2\pi)^{3/2}} \int \frac{d\omega}{(2\pi)^{1/2}} \rho(\mathbf{k}, \omega) e^{-i\omega t + i\mathbf{k} \cdot \mathbf{x}}. \quad (2.4)$$

With the relativistic volume elements $d^4x = d^3\mathbf{x} dx^0$ and $d^4k = d^3\mathbf{k} dk^0$, we can write these equations equivalently as

$$\rho(k) = \int \frac{d^4x}{(2\pi)^2} \rho(x) e^{-ikx}, \quad (2.5)$$

$$\rho(x) = \int \frac{d^4k}{(2\pi)^2} \rho(k) e^{ikx}. \quad (2.6)$$

In particular, under Fourier transformation ∂_μ is mapped to ik_μ . For a response relation as

$$\rho(x) = \int d^4x' \chi(x, x') \varphi(x'), \quad (2.7)$$

we require “covariance under Fourier transformation”, i.e., the Fourier transformed quantities should obey the analogous relation

$$\rho(k) = \int d^4k' \chi(k, k') \varphi(k'). \quad (2.8)$$

This implies that the response function transforms as

$$\chi(k, k') = \int \frac{d^4x}{(2\pi)^2} \int \frac{d^4x'}{(2\pi)^2} e^{-ikx} \chi(x, x') e^{ik'x'}, \quad (2.9)$$

$$\chi(x, x') = \int \frac{d^4k}{(2\pi)^2} \int \frac{d^4k'}{(2\pi)^2} e^{ikx} \chi(k, k') e^{-ik'x'}. \quad (2.10)$$

Our conventions are in accord with a functional chain rule,

$$\chi(k, k') = \frac{\delta\rho(k)}{\delta\varphi(k')} = \int d^4x \int d^4x' \frac{\delta\rho(k)}{\delta\rho(x)} \frac{\delta\rho(x)}{\delta\varphi(x')} \frac{\delta\varphi(x')}{\delta\varphi(k')} \quad (2.11)$$

$$= \int d^4x \int d^4x' \frac{e^{-ikx}}{(2\pi)^2} \chi(x, x') \frac{e^{ik'x'}}{(2\pi)^2}. \quad (2.12)$$

The above general formulae may simplify on physical grounds: we always assume *homogeneity in time* and sometimes also *homogeneity in space*. The response function can then be written as

$$\chi(x, x') = \chi(\mathbf{x} - \mathbf{x}', t - t'), \quad (2.13)$$

or in the Fourier domain

$$\chi(k, k') = c \chi(\mathbf{k}, \omega) \delta^3(\mathbf{k} - \mathbf{k}') \delta(\omega - \omega'). \quad (2.14)$$

With $\mathbf{r} = \mathbf{x} - \mathbf{x}'$ and $\tau = t - t'$, the Fourier transformation then reads

$$\chi(\mathbf{k}, \omega) = c \int d^3\mathbf{r} \int d\tau \chi(\mathbf{r}, \tau) e^{i\omega\tau - i\mathbf{k}\cdot\mathbf{r}}, \quad (2.15)$$

$$\chi(\mathbf{r}, \tau) = \frac{1}{c} \int \frac{d^3\mathbf{k}}{(2\pi)^3} \int \frac{d\omega}{2\pi} \chi(\mathbf{k}, \omega) e^{-i\omega\tau + i\mathbf{k}\cdot\mathbf{r}}. \quad (2.16)$$

In particular, we note that the response function $\chi(\mathbf{k}, \omega)$ does not Fourier transform in the same way as the field quantity $\rho(\mathbf{k}, \omega)$ (compare Eqs. (2.3) and (2.15)). Furthermore, the relation

$$\rho(\mathbf{x}, t) = c \int d^3 \mathbf{x}' \int dt' \chi(\mathbf{x} - \mathbf{x}', t - t') \varphi(\mathbf{x}', t') \quad (2.17)$$

is equivalent to

$$\rho(\mathbf{k}, \omega) = \chi(\mathbf{k}, \omega) \varphi(\mathbf{k}, \omega), \quad (2.18)$$

which shows that the Fourier covariance is lost if one works with the reduced transform $\chi(\mathbf{k}, \omega)$. Finally, we note that in the limit $\omega \rightarrow 0$, $\chi(\mathbf{k}, 0)$ relates the *static* parts $\rho(\mathbf{k}, 0)$ and $\varphi(\mathbf{k}, 0)$ of the respective field quantities, whereas the response function itself

$$\chi(\mathbf{x} - \mathbf{x}', t - t') = \frac{1}{c} \delta(t - t') \int \frac{d^3 \mathbf{k}}{(2\pi)^3} \chi(\mathbf{k}, 0) e^{i\mathbf{k} \cdot (\mathbf{x} - \mathbf{x}')} \quad (2.19)$$

is not static (time-independent) but *instantaneous*.

Projections and rotations. Longitudinal and transverse projection operators are defined in Fourier space by

$$(P_L)_{ij}(\mathbf{k}) = \frac{k_i k_j}{|\mathbf{k}|^2}, \quad (2.20)$$

$$(P_T)_{ij}(\mathbf{k}) = \delta_{ij} - \frac{k_i k_j}{|\mathbf{k}|^2}. \quad (2.21)$$

Any vector field $\mathbf{E}(\mathbf{k})$ can be decomposed into its longitudinal and transverse parts

$$\mathbf{E}(\mathbf{k}) = \overset{\leftrightarrow}{P}_L(\mathbf{k}) \mathbf{E}(\mathbf{k}) + \overset{\leftrightarrow}{P}_T(\mathbf{k}) \mathbf{E}(\mathbf{k}) \equiv \mathbf{E}_L(\mathbf{k}) + \mathbf{E}_T(\mathbf{k}), \quad (2.22)$$

where

$$\mathbf{E}_L(\mathbf{k}) = \frac{\mathbf{k}(\mathbf{k} \cdot \mathbf{E}(\mathbf{k}))}{|\mathbf{k}|^2}, \quad (2.23)$$

and

$$\mathbf{E}_T(\mathbf{k}) = \frac{|\mathbf{k}|^2 \mathbf{E}(\mathbf{k}) - \mathbf{k}(\mathbf{k} \cdot \mathbf{E}(\mathbf{k}))}{|\mathbf{k}|^2} = -\frac{\mathbf{k} \times (\mathbf{k} \times \mathbf{E}(\mathbf{k}))}{|\mathbf{k}|^2}. \quad (2.24)$$

These two parts are orthogonal with respect to the euclidean inner product,

$$\mathbf{E}_L^*(\mathbf{k}) \cdot \mathbf{E}_T(\mathbf{k}) = 0. \quad (2.25)$$

In real space, transverse vector fields are divergence free, $\nabla \cdot \mathbf{E}_T(\mathbf{x}) = 0$, whereas longitudinal vector fields can be represented as the gradient of a scalar function, $\mathbf{E}_L(\mathbf{x}) = -\nabla\varphi(\mathbf{x})$. They are orthogonal with respect to the inner product

$$\int d^3\mathbf{x} \mathbf{E}_L(\mathbf{x}) \cdot \mathbf{E}_T(\mathbf{x}) = 0 \quad (2.26)$$

(where we assume that $\mathbf{E}(\mathbf{x})$ is real), and the decomposition (2.22) translates into the Helmholtz vector theorem

$$\mathbf{E}(\mathbf{x}) = -\frac{1}{4\pi}\nabla \int d^3\mathbf{x}' \frac{(\nabla' \cdot \mathbf{E})(\mathbf{x}')}{|\mathbf{x} - \mathbf{x}'|} + \frac{1}{4\pi}\nabla \times \int d^3\mathbf{x}' \frac{(\nabla' \times \mathbf{E})(\mathbf{x}')}{|\mathbf{x} - \mathbf{x}'|}. \quad (2.27)$$

In addition to the longitudinal and transverse projectors, we define the *transverse rotation operator* (cf. [51, Eq. (A.3)])

$$(R_T)_{ij}(\mathbf{k}) = \epsilon_{ij\ell} \frac{k_\ell}{|\mathbf{k}|}, \quad (2.28)$$

which acts on a vector field $\mathbf{E}(\mathbf{k})$ by

$$\overset{\leftrightarrow}{R}_T(\mathbf{k}) \mathbf{E}(\mathbf{k}) = \frac{\mathbf{k} \times \mathbf{E}(\mathbf{k})}{|\mathbf{k}|}. \quad (2.29)$$

The three vectors $P_L \mathbf{E}$, $P_T \mathbf{E}$ and $R_T \mathbf{E}$ are orthogonal, and the operators P_L , P_T and R_T generate an algebra with the multiplication table shown in Table 1.

2.2. Fields, potentials and sources

The Maxwell equations relate the electric and magnetic fields $\{\mathbf{E}, \mathbf{B}\}$ to the charge and current densities $\{\rho, \mathbf{j}\}$ as

$$\nabla \cdot \mathbf{E} = \rho/\varepsilon_0, \quad (2.30)$$

$$\nabla \times \mathbf{B} - \frac{1}{c^2} \frac{\partial \mathbf{E}}{\partial t} = \mu_0 \mathbf{j}, \quad (2.31)$$

$$\nabla \cdot \mathbf{B} = 0, \quad (2.32)$$

$$\nabla \times \mathbf{E} + \frac{\partial \mathbf{B}}{\partial t} = 0, \quad (2.33)$$

\cdot	\overleftrightarrow{P}_L	\overleftrightarrow{P}_T	\overleftrightarrow{R}_T
\overleftrightarrow{P}_L	\overleftrightarrow{P}_L	0	0
\overleftrightarrow{P}_T	0	\overleftrightarrow{P}_T	\overleftrightarrow{R}_T
\overleftrightarrow{R}_T	0	\overleftrightarrow{R}_T	$-\overleftrightarrow{P}_T$

Table 1: Multiplication table for the longitudinal projection operator $P_L(\mathbf{k})$, the transverse projection operator $P_T(\mathbf{k})$ and the transverse rotation operator $R_T(\mathbf{k})$. These operators acting in the three-dimensional euclidean space are defined by Eqs. (2.20), (2.21) and (2.28).

where $c^2 = 1/(\varepsilon_0\mu_0)$. These equations are also referred to as Gauss's law, Ampère's law (with Maxwell's correction), Gauss's law for magnetism and Faraday's law, respectively. There is a general consensus that these equations are universally valid on a microscopic scale [1, 6, 11, 79–82]. In the context of electrodynamics of media, see in particular the remarks in Ref. [5, pp. 3 f.]. The sources on the right hand side of Eqs. (2.30) and (2.31) have to satisfy the continuity equation on grounds of consistency,

$$\frac{\partial \rho}{\partial t} + \nabla \cdot \mathbf{j} = 0. \quad (2.34)$$

Electromagnetic potentials $\{\varphi, \mathbf{A}\}$ are introduced by

$$\mathbf{E} = -\nabla\varphi - \frac{\partial \mathbf{A}}{\partial t}, \quad (2.35)$$

$$\mathbf{B} = \nabla \times \mathbf{A}. \quad (2.36)$$

These are only determined up to gauge transformations: two potentials $\{\varphi, \mathbf{A}\}$ and $\{\varphi', \mathbf{A}'\}$ (defined on the whole three-dimensional space) lead to the same electric and magnetic fields $\{\mathbf{E}, \mathbf{B}\}$ if and only if they differ by a pure gauge, i.e., if there is a real function f such that

$$\varphi' = \varphi - \frac{\partial f}{\partial t}, \quad (2.37)$$

$$\mathbf{A}' = \mathbf{A} + \nabla f. \quad (2.38)$$

A relativistic formulation of electrodynamics is obtained by introducing the four-current density

$$j^\mu = (c\rho, j_1, j_2, j_3)^T, \quad (2.39)$$

the four-potential

$$A^\mu = (\varphi/c, A_1, A_2, A_3)^T, \quad (2.40)$$

and the field strength tensor (cf. [81], which also uses the metric convention (2.1)),

$$F^{\mu\nu} = \begin{pmatrix} 0 & E_1/c & E_2/c & E_3/c \\ -E_1/c & 0 & B_3 & -B_2 \\ -E_2/c & -B_3 & 0 & B_1 \\ -E_3/c & B_2 & -B_1 & 0 \end{pmatrix}. \quad (2.41)$$

The electric and magnetic fields can be gained back by from the field strength tensor by

$$E_i = cF^{0i}, \quad (2.42)$$

$$B_i = \frac{1}{2}\epsilon_{ik\ell}F^{k\ell}. \quad (2.43)$$

In a relativistic notation, the continuity equation (2.34) can be written as

$$\partial_\mu j^\mu = 0, \quad (2.44)$$

while the gauge tranformation (2.37)–(2.38) reads

$$A'^\mu = A^\mu + \partial^\mu f. \quad (2.45)$$

The Maxwell equations (2.30)–(2.33) can also be written in a manifestly covariant form, see [12, Section 2.3].

2.3. Functional interdependencies

We now discuss in how far any electromagnetic system can be described equivalently by its four-current j^μ , by its four-potential A^μ , or by its field strength tensor $F^{\mu\nu}$. For this purpose, we relate any of these quantities to the remaining two (see Fig. 1): Given the four-potential, the field strength tensor is determined by

$$F^{\mu\nu}[A^\lambda] = \partial^\mu A^\nu - \partial^\nu A^\mu = (\eta^\nu_\lambda \partial^\mu - \eta^\mu_\lambda \partial^\nu) A^\lambda, \quad (2.46)$$

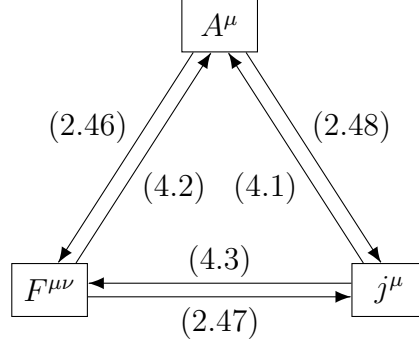


Figure 1: Mutual relationships between the electromagnetic four-potential A^μ , the field strength tensor $F^{\mu\nu}$ and the four-current density j^μ . The arrow labels refer to the equation numbers in the text.

with $\eta^\mu{}_\nu = \eta^{\mu\alpha}\eta_{\alpha\nu} = \text{diag}(1, 1, 1, 1)$. This formula is equivalent to the defining equations (2.35)–(2.36) for the potentials.

Given the field strength tensor, the four-current is determined by the inhomogeneous Maxwell equations, which read in a manifestly covariant form

$$j^\mu[F^{\nu\lambda}] = \frac{1}{\mu_0} \partial_\lambda F^{\mu\lambda} = \frac{1}{2\mu_0} (\eta^\mu{}_\nu \partial_\lambda - \eta^\mu{}_\lambda \partial_\nu) F^{\nu\lambda}. \quad (2.47)$$

Combining Eqs. (2.46) and (2.47), we further obtain the four-current in terms of the four-potential,

$$j^\mu[A^\nu] = \frac{1}{\mu_0} (\eta^\mu{}_\nu \square + \partial^\mu \partial_\nu) A^\nu, \quad (2.48)$$

where

$$\square = -\partial_\lambda \partial^\lambda = \frac{1}{c^2} \frac{\partial^2}{\partial t^2} - \Delta \quad (2.49)$$

is called the d'Alembert operator. Eq. (2.48) can also be regarded as the equation of motion for the four-potential in terms of the four-current. The solution to this differential equation yields the four-potential $A^\mu[j^\nu]$ in terms of the sources. Expressing the sources in terms of the field strength tensor then leads to the four-potential $A^\mu[F^{\nu\lambda}]$ as a functional of the field strength tensor. We will construct the most general solution to the differential equation (2.48) and derive explicit expressions for the functionals $A^\mu[j^\nu]$ and $A^\mu[F^{\nu\lambda}]$ in the following sections 3 and 4.

It remains to express the electric and magnetic fields as functionals of the sources, which is equivalent to finding the general solution of the Maxwell equations. By applying the d'Alembert operator on both sides of Eq. (2.46) and using Eq. (2.48), we obtain the wave equation for the components of the field strength tensor,

$$\square F^{\mu\nu} = \mu_0(\partial^\mu j^\nu - \partial^\nu j^\mu). \quad (2.50)$$

This is equivalent to the equations of motion for the decoupled electric and magnetic fields in terms of the charge and current densities,

$$\square \mathbf{E} = -\frac{1}{\varepsilon_0} \nabla \rho - \mu_0 \frac{\partial \mathbf{j}}{\partial t}, \quad (2.51)$$

$$\square \mathbf{B} = \mu_0 \nabla \times \mathbf{j}. \quad (2.52)$$

The solutions to these equations formally yield the functional $F^{\mu\nu}[j^\lambda]$. This, however, requires a more precise discussion for the following reasons: (i) The wave equations require initial conditions to be fixed, and (ii) they follow from the Maxwell equations, but are not equivalent to them. This means, there are solutions to the wave equations which do not obey the Maxwell equations. The initial value problem for both the wave equation and the Maxwell equations will therefore be discussed in the next section.

3. Electromagnetic Green functions

3.1. Initial value problem for scalar wave equation

In order to determine the functional $F^{\mu\nu}[j^\lambda]$ explicitly from Eq. (2.50), we first study the initial value problem of the inhomogeneous scalar wave equation

$$\square \phi(x) = \mu_0 g(x). \quad (3.1)$$

Thus, we seek the uniquely determined solution to this equation with given initial conditions

$$\phi(t_0, \mathbf{x}) = \phi_0(\mathbf{x}), \quad (3.2)$$

$$(\partial_t \phi)(t_0, \mathbf{x}) = \phi_1(\mathbf{x}). \quad (3.3)$$

For this purpose, we introduce the *scalar Green function* \mathbb{D}_0 and the *propagator* \mathcal{U}_0 . The former is a distribution obeying

$$\square \mathbb{D}_0(x - x') = \mu_0 \delta^4(x - x') \equiv \frac{\mu_0}{c} \delta^3(\mathbf{x} - \mathbf{x}') \delta(t - t'), \quad (3.4)$$

whereas the latter is subject to

$$\square \mathcal{U}_0(x - x') = 0 \quad (3.5)$$

and the initial conditions

$$\mathcal{U}_0(\mathbf{x} - \mathbf{x}', \tau = 0) = 0, \quad (3.6)$$

$$\frac{1}{c} (\partial_t \mathcal{U}_0)(\mathbf{x} - \mathbf{x}', \tau = 0) = \delta^3(\mathbf{x} - \mathbf{x}'), \quad (3.7)$$

where $\tau = t - t'$. The equation (3.4) for the Green function has infinitely many solutions, the most important of which are the *retarded* Green function \mathbb{D}^+ and the *advanced* Green function \mathbb{D}^- . These are given by

$$\mathbb{D}_0^\pm(\mathbf{x} - \mathbf{x}', t - t') = \frac{\mu_0}{4\pi c} \frac{\delta(t - t' \mp |\mathbf{x} - \mathbf{x}'|/c)}{|\mathbf{x} - \mathbf{x}'|}, \quad (3.8)$$

or in the Fourier domain

$$\mathbb{D}_0^\pm(\mathbf{k}, \omega) = \frac{\mu_0}{-(\omega/c \pm i\eta)^2 + |\mathbf{k}|^2} \equiv \frac{\mu_0}{k^2}, \quad (3.9)$$

with $\eta \rightarrow 0^+$. (The infinitesimal η will be suppressed in the sequel.) With these definitions, the unique solution to the equations (3.5)–(3.7) for the propagator can be written as (cf. [66, Eq. (8.34)])

$$\mathcal{U}_0 = \frac{1}{\mu_0} (\mathbb{D}_0^+ - \mathbb{D}_0^-). \quad (3.10)$$

The general solution of the inhomogeneous wave equation (3.1) with initial conditions (3.2)–(3.3) is now given by the Duhamel formula (cf. [83, Section 1.3] and [84, Section 3.3])

$$\begin{aligned} \phi(\mathbf{x}, t) &= c \int_{t_0}^{\infty} dt' \int d^3\mathbf{x}' \mathbb{D}_0^+(\mathbf{x} - \mathbf{x}', t - t') g(\mathbf{x}', t') \\ &\quad + \frac{1}{c} \int d^3\mathbf{x}' (\partial_t \mathcal{U}_0)(\mathbf{x} - \mathbf{x}', t - t_0) \phi_0(\mathbf{x}', t_0) \\ &\quad + \frac{1}{c} \int d^3\mathbf{x}' \mathcal{U}_0(\mathbf{x} - \mathbf{x}', t - t_0) \phi_1(\mathbf{x}', t_0). \end{aligned} \quad (3.11)$$

Note that the field ϕ naturally decomposes into a retarded part and a vacuum solution, $\phi = \phi_{\text{ret}} + \phi_{\text{vac}}$, which satisfy $\square\phi_{\text{ret}} = \mu_0 g$ and $\square\phi_{\text{vac}} = 0$, respectively. The retarded part is given in terms of the sources by the retarded Green function, while the vacuum part is given in terms of the initial conditions by the propagator.

3.2. Initial value problem for Maxwell equations

Formally, the decoupled equations (2.51)–(2.52) for the electric and magnetic fields $\{\mathbf{E}, \mathbf{B}\}$ are inhomogeneous wave equations. In principle, the corresponding initial value problem can therefore be solved by the method of the previous subsection. Concretely, this means that the electric and magnetic fields are uniquely determined by the formula (3.11) once we are given appropriate initial conditions for these fields and their first time-derivatives. However, in the case of the Maxwell equations, the initial value problem is more complicated due to the fact that these initial conditions cannot be prescribed arbitrarily: generally, the initial conditions at time $t = t_0$ have to be given such that they fulfill the equations (cf. [83, p. 29])

$$\nabla \cdot \mathbf{E}_0 = \rho_0 / \varepsilon_0, \quad (3.12)$$

$$\nabla \times \mathbf{E}_0 = -(\partial_t \mathbf{B})_0, \quad (3.13)$$

$$\nabla \cdot \mathbf{B}_0 = 0, \quad (3.14)$$

$$\nabla \times \mathbf{B}_0 = \mu_0 \mathbf{j}_0 + (\partial_t \mathbf{E})_0 / c^2, \quad (3.15)$$

where $\mathbf{E}_0(\mathbf{x}) = \mathbf{E}(\mathbf{x}, t_0)$, $(\partial_t \mathbf{E})_0(\mathbf{x}) = (\partial_t \mathbf{E})(\mathbf{x}, t_0)$, etc. These equations are formally identical to the Maxwell equations evaluated at time $t = t_0$, but they have the meaning of *constraints*, namely, they constrain the freedom of choosing initial conditions for the Maxwell equations. Any solution $\{\mathbf{E}(\mathbf{x}, t), \mathbf{B}(\mathbf{x}, t)\}$ of the inhomogeneous wave equations (2.51)–(2.52) with initial conditions at time $t = t_0$ fulfilling Eqs. (3.12)–(3.15) is also a solution of the Maxwell equations for all later times $t > t_0$. In particular, the above constraints imply that only the transverse fields \mathbf{E}_T and \mathbf{B} can be prescribed at the initial time. The longitudinal part of the magnetic field has to vanish, while the longitudinal part of the electric field is completely determined in terms of the charges and therefore does not carry *independent degrees of freedom*. Furthermore, the initial condition for $\partial_t \mathbf{B}$ is determined by the initial condition for \mathbf{E}_T , and the initial condition for $\partial_t \mathbf{E}$ is determined by the initial condition for \mathbf{B} and the spatial current.

These consideration show that the electromagnetic fields are in general not uniquely determined by the sources and therefore do not constitute functionals of the sources alone. Instead, they also depend on the initial conditions. Fortunately, when we deal with electromagnetic perturbations of a sample which are produced in the laboratory, we can assume that all observable (external and induced) quantities, i.e., the charge and current densities as well as the corresponding fields, vanish before some initial switching-on. Thus, if we choose t_0 sufficiently early (before the external perturbation is switched on) we are entitled to choose identically vanishing initial conditions $\mathbf{E}_0 \equiv 0$, $(\partial_t \mathbf{E})_0 \equiv 0$, etc. The solutions of the wave equations (2.50) or (2.51)–(2.52) can then be deduced componentwise in terms of the respective sources. In particular, we obtain the functional

$$F^{\mu\nu}[j^\lambda] = \mathbb{D}_0 (\partial^\mu j^\nu - \partial^\nu j^\mu) \quad (3.16)$$

$$= (\partial^\mu \mathbb{D}_0 \eta^\nu{}_\lambda - \partial^\nu \mathbb{D}_0 \eta^\mu{}_\lambda) j^\lambda, \quad (3.17)$$

where we have used partial integration, and where

$$\mu_0 \square^{-1} = \mathbb{D}_0 \equiv \mathbb{D}_0^+ \quad (3.18)$$

denotes the retarded Green function of the scalar wave equation. We stress that this choice of the Green function is dictated by the underlying initial-value problem. The electromagnetic fields defined in this way are uniquely determined by the sources and hence constitute functionals of them.

3.3. Tensorial Green function

We now turn to the explicit functional $A^\mu[j^\nu]$ for the potentials in terms of the sources. Consider the equation of motion (2.48) of the four-potential in terms of the four-current,

$$(\eta^\mu{}_\nu \square + \partial^\mu \partial_\nu) A^\nu = \mu_0 j^\mu. \quad (3.19)$$

Again we assume that the initial conditions for the electric and magnetic fields vanish, and hence we seek a *tensorial Green function* $(D_0)^\mu{}_\nu$ for the four-potential that is *retarded*, i.e.,

$$(D_0)^\mu{}_\nu(\mathbf{x} - \mathbf{x}', t - t') = 0 \quad \text{if } t < t'. \quad (3.20)$$

Given the current j^μ , the Green function should yield by

$$A^\nu = (D_0)^\nu{}_\mu j^\mu \quad (3.21)$$

a solution A^ν to the equation of motion (3.19). The general solution to this inhomogeneous differential equation can then be obtained by adding to the particular solution (3.21) the general solution of the corresponding homogeneous differential equation. With vanishing initial fields, the solutions of the homogeneous equation are precisely given by the *pure gauges*,

$$A^\nu = \partial^\nu f, \quad (3.22)$$

as

$$(\eta^\mu{}_\nu \square + \partial^\mu \partial_\nu) \partial^\nu f = 0. \quad (3.23)$$

We will now first construct the most general form of a tensorial Green function, and in the following subsection discuss special expressions which are obtained by gauge fixing conditions.

In analogy to the scalar case, one could expect that the tensorial Green function satisfies the equation

$$(\eta^\mu{}_\lambda \square + \partial^\mu \partial_\lambda) (D_0)^\lambda{}_\nu (x - x') \stackrel{?}{=} \eta^\mu{}_\nu \delta^4(x - x'). \quad (3.24)$$

However, it turns out that this equation has no solutions (see [79, Section 3.4] and [85, Section 4.3]). The key to overcome this problem is to restrict oneself to *physical* four-currents which satisfy the continuity equation $\partial_\mu j^\mu = 0$. Hence, only for such currents the Green function should yield by Eq. (3.21) a four-potential which solves the equation of motion (3.19). To put this program into practice, we define projection operators on the Minkowski longitudinal and transverse parts in Fourier space by (cf. [85, p. 36])

$$(P_L)^\mu{}_\nu(k) = \frac{k^\mu k_\nu}{k^2}, \quad (3.25)$$

$$(P_T)^\mu{}_\nu(k) = \eta^\mu{}_\nu - \frac{k^\mu k_\nu}{k^2}, \quad (3.26)$$

with $k^2 = k_\mu k^\mu$. (These projection operators must not be confused with their euclidean counterparts introduced in Section 2.1.) The Minkowski projection operators are orthogonal with respect to the Minkowski inner product, and hence any vector field $C^\mu(k)$ can be decomposed into its longitudinal and transverse parts $(C_L)^\mu = (P_L)^\mu{}_\nu C^\nu$ and $(C_T)^\mu = (P_T)^\mu{}_\nu C^\nu$, such that

$$(C_L)^\mu + (C_T)^\mu = C^\mu, \quad (3.27)$$

$$(C_L)_\mu (C_T)^\mu = 0. \quad (3.28)$$

In real space, transverse vector fields have vanishing four-divergence, $\partial_\mu (C_T)^\mu(x) = 0$, whereas longitudinal vector fields can be represented as a four-gradient $(C_L)^\mu(x) = (\partial^\mu f)(x)$. With these definitions, the continuity equation is equivalent to the statement that j^μ is a Minkowski-transverse four-vector, i.e., $P_L j = 0$ and $P_T j = j$. Moreover, the equation of motion (3.19) can be written in Fourier space as

$$k^2 (P_T)^\mu{}_\nu(k) A^\nu(k) = \mu_0 j^\mu(k), \quad (3.29)$$

or in an abridged notation

$$k^2 P_T A = \mu_0 j. \quad (3.30)$$

Putting the ansatz (3.21) into this equation leads to the condition

$$k^2 P_T D_0 j = \mu_0 j, \quad (3.31)$$

which should hold for any physical (Minkowski-transverse) four-current j . This implies the *defining equation* for the free tensorial Green function:

$$k^2 P_T D_0 P_T = \mu_0 P_T. \quad (3.32)$$

In order to construct the most general solution to this equation, we note that any tensorial operator D_0 can be decomposed into four independent parts as

$$D_0 = P_T D_0 P_T + P_L D_0 P_T + P_T D_0 P_L + P_L D_0 P_L, \quad (3.33)$$

and these four parts contain $3 \times 3 = 9$, $1 \times 3 = 3$, $3 \times 1 = 3$ and $1 \times 1 = 1$ independent component functions, respectively. Evidently, Eq. (3.32) fixes only the first operator as

$$P_T D_0 P_T = \frac{\mu_0}{k^2} P_T, \quad (3.34)$$

whereas the remaining operators can be chosen arbitrarily. We note that this solution is only formal in that it involves the singular expression $1/k^2$, the inverse Fourier transform of which is not well-defined. (The same problem arises if one tries to define the Minkowski projection operators in real space.) In accordance with our previous considerations, we interpret any contribution of the form $1/k^2$ as a retarded Green function in the sense of Eq. (3.9), hence

$$P_T D_0 P_T = \mathbb{D}_0 P_T. \quad (3.35)$$

We conclude that in constructing the most general tensorial Green function $(D_0)^\mu{}_\nu$, one may choose three arbitrary operators on the right hand side of Eq. (3.33), whereas the first contribution is fixed by Eq. (3.35), with $\mathbb{D}_0 = \mu_0 \square^{-1}$ being the retarded Green function of the scalar wave equation.

An explicit expression for the tensorial Green function is now given by

$$(D_0)^\mu{}_\nu(k) = \mathbb{D}_0(k) \left(\eta^\mu{}_\nu - \frac{k^\mu k_\nu}{k^2} \right) + k^\mu \tilde{f}_\nu(k) + \tilde{g}^\mu(k) k_\nu + k^\mu \tilde{h}(k) k_\nu, \quad (3.36)$$

where the functions \tilde{f}_ν , \tilde{g}^μ and \tilde{h} can be chosen arbitrarily up to the constraints of Minkowski-transversality

$$\tilde{f}_\nu(k) k^\nu = k_\mu \tilde{g}^\mu(k) = 0. \quad (3.37)$$

Conversely, these functions can be reconstructed from a given tensorial Green function by (suppressing momentum dependencies in the notation)

$$\tilde{f}_\nu(k) = \frac{k_\lambda}{k^2} (D_0)^\lambda{}_\rho (P_T)^\rho{}_\nu, \quad (3.38)$$

$$\tilde{g}^\mu(k) = (P_T)^\mu{}_\lambda (D_0)^\lambda{}_\rho \frac{k^\rho}{k^2}, \quad (3.39)$$

$$\tilde{h}(k) = \frac{k_\lambda}{k^2} (D_0)^\lambda{}_\rho \frac{k^\rho}{k^2}. \quad (3.40)$$

In order to obtain a more symmetric form of Eq. (3.36), we rewrite it in terms of dimensionless parameter functions f_ν , g^μ and h as

$$(D_0)^\mu{}_\nu(k) = \mathbb{D}_0(k) \left(\eta^\mu{}_\nu + \frac{ck^\mu}{\omega} f_\nu(k) + g^\mu(k) \frac{ck_\nu}{\omega} + \frac{ck^\mu}{\omega} h(k) \frac{ck_\nu}{\omega} \right). \quad (3.41)$$

These new functions are related to the previously defined functions by

$$f_\nu(k) = \frac{\omega}{c} \frac{k^2}{\mu_0} \tilde{f}_\nu(k), \quad (3.42)$$

$$g^\mu(k) = \frac{\omega}{c} \frac{k^2}{\mu_0} \tilde{g}^\mu(k), \quad (3.43)$$

$$h(k) = \frac{\omega^2}{c^2} \left(\frac{k^2}{\mu_0} \tilde{h}(k) - \frac{1}{k^2} \right). \quad (3.44)$$

Again, f_ν and g^μ are Minkowski-transverse in the sense that

$$f_\nu(k) k^\nu = k_\mu g^\mu(k) = 0, \quad (3.45)$$

and consequently they can be written as

$$f_\nu(\mathbf{k}, \omega) = \left(-\frac{c\mathbf{k} \cdot \mathbf{f}(\mathbf{k}, \omega)}{\omega}, \mathbf{f}(\mathbf{k}, \omega) \right), \quad (3.46)$$

$$g^\mu(\mathbf{k}, \omega) = \left(\frac{c\mathbf{k} \cdot \mathbf{g}(\mathbf{k}, \omega)}{\omega}, \mathbf{g}(\mathbf{k}, \omega) \right)^T. \quad (3.47)$$

Our formula (3.41) for the tensorial Green function generalizes the formulae of Ref. [86, Section VIII. §77]. The seven dimensionless functions \mathbf{f} , \mathbf{g} and h can be chosen arbitrarily, and for each choice of them we obtain by Eq. (3.41) a tensorial electromagnetic Green function which satisfies Eq. (3.32).

3.4. Gauge fixing

Special forms of the tensorial Green function correspond to special choices of the free parameter functions in Eq. (3.41). For example, the *Lorenz Green function* (cf. [87, Eq. (41) with $\alpha = 0$]) reads

$$(D_0)^\mu{}_\nu(k) = \mathbb{D}_0(k) (P_T)^\mu{}_\nu(k), \quad (3.48)$$

and is obtained from

$$\mathbf{f}(k) = \mathbf{g}(k) = 0, \quad h(k) = -\frac{\omega^2}{c^2 k^2}. \quad (3.49)$$

With this definition, $A^\mu = (D_0)^\mu{}_\nu j^\nu$ implies the Lorenz gauge condition [87]

$$\partial_\mu A^\mu = 0. \quad (3.50)$$

In relativistic quantum field theory, one mainly uses the *Feynman Green function* (cf. [87, Eq. (41) with $\alpha = 1$] and [86, Eq. (77.8)]),

$$(D_0)^\mu{}_\nu(k) = \mathbb{D}_0(k) \eta^\mu{}_\nu, \quad (3.51)$$

which corresponds to $\mathbf{f}(k) = \mathbf{g}(k) = h(k) = 0$. By contrast, in non-relativistic quantum field theory the *Coulomb Green function* [86, Eqs. (77.12)–(77.13)]

$$(D_0)^\mu{}_\nu(\mathbf{k}, \omega) = \begin{pmatrix} \mu_0/|\mathbf{k}|^2 & 0 \\ 0 & \mathbb{D}_0(\mathbf{k}, \omega) \overset{\leftrightarrow}{P}_T(\mathbf{k}) \end{pmatrix} \quad (3.52)$$

is frequently used. This requires the more involved choice

$$\mathbf{f}(\mathbf{k}, \omega) = \mathbf{g}(\mathbf{k}, \omega) = \frac{\omega c \mathbf{k}}{c^2 |\mathbf{k}|^2 - \omega^2} \frac{\omega^2}{c^2 |\mathbf{k}|^2}, \quad (3.53)$$

$$h(\mathbf{k}, \omega) = -\frac{\omega^2}{c^2 |\mathbf{k}|^2 - \omega^2} \left(1 + \frac{\omega^2}{c^2 |\mathbf{k}|^2} \right). \quad (3.54)$$

It implies the Coulomb gauge of the electromagnetic potential $A = D_0 j$, which reads

$$\nabla \cdot \mathbf{A} = 0. \quad (3.55)$$

Finally, for electrodynamics of materials the *temporal Green function*

$$(D_0)^\mu{}_\nu(\mathbf{k}, \omega) = \mathbb{D}_0(\mathbf{k}, \omega) \begin{pmatrix} 0 & 0 \\ -c\mathbf{k}/\omega & \overset{\leftrightarrow}{1} \end{pmatrix} \quad (3.56)$$

turns out to be particularly useful. This is obtained from

$$\mathbf{f}(\mathbf{k}, \omega) = \frac{\omega c \mathbf{k}}{c^2 |\mathbf{k}|^2 - \omega^2}, \quad (3.57)$$

$$\mathbf{g}(\mathbf{k}, \omega) = 0, \quad (3.58)$$

$$h(\mathbf{k}, \omega) = -\frac{\omega^2}{c^2 |\mathbf{k}|^2 - \omega^2}. \quad (3.59)$$

In this case the electromagnetic potential satisfies

$$A^0 \equiv \varphi/c = 0, \quad (3.60)$$

which is the temporal gauge condition. In the following, we will employ this last gauge condition, because it facilitates the expression of the vector potential \mathbf{A} in terms of the electric and magnetic fields $\{\mathbf{E}, \mathbf{B}\}$ (see Sections 4.1 and 4.2).

4. Canonical functional

4.1. Temporal gauge

We now come back to the problem of finding explicit expressions for the four-potential as a functional of the electric and magnetic fields, $A^\mu =$

$A^\mu[F^{\nu\lambda}]$. The tensorial Green function being fixed, the general solution to the equation of motion for the four-potential can be written as

$$A^\mu[j^\nu] = (D_0)^\mu{}_\nu j^\nu + \partial^\mu f, \quad (4.1)$$

with an arbitrary pure gauge $\partial^\mu f$. In particular, this means that the four-potential is determined in terms of the four-current only up to gauge transformations. Combining Eq. (4.1) with Eq. (2.47) and by partial integration, the four-potential can be expressed in terms of the field strength tensor as

$$A^\mu[F^{\nu\lambda}] = \frac{1}{2\mu_0} (\partial_\lambda (D_0)^\mu{}_\nu - \partial_\nu (D_0)^\mu{}_\lambda) F^{\nu\lambda} + \partial^\mu f. \quad (4.2)$$

On the other hand, combining Eq. (4.1) with Eq. (2.46) yields the fields in terms of the sources,

$$F^{\mu\nu}[j^\lambda] = (\partial^\mu (D_0)^\nu{}_\lambda - \partial^\nu (D_0)^\mu{}_\lambda) j^\lambda. \quad (4.3)$$

This last functional does not depend on the choice of the tensorial Green function, and by taking the Feynman Green function (3.51) one sees that it is equivalent to Eq. (3.17). Hence it corresponds to the retarded solution of Maxwell's equations with vanishing initial conditions.

We now rewrite the functional (4.2) explicitly in terms of the electric and magnetic fields using the temporal gauge condition (3.60). First note that $\varphi = 0$ implies

$$\mathbf{E} = -\partial_t \mathbf{A}, \quad (4.4)$$

$$\mathbf{B} = \nabla \times \mathbf{A}. \quad (4.5)$$

Plugging these equations into the Maxwell equations, we find the equation of motion for the vector potential in the temporal gauge,

$$\square \mathbf{A}(\mathbf{x}, t) = \mu_0 \mathbf{j}(\mathbf{x}, t) + \int_{t_0}^t dt' \nabla \rho(\mathbf{x}, t') / \varepsilon_0. \quad (4.6)$$

Here, the lower bound t_0 of the time integral is arbitrary: alternating the lower bound modifies the vector potential by a time-independent gradient, which has no bearing on the electromagnetic fields. Solving Eq. (4.6) for the vector potential by means of the scalar Green function $\mathbb{D}_0 = \mu_0 \square^{-1}$ yields in Fourier space

$$\mathbf{A}(\mathbf{k}, \omega) = \mathbb{D}_0(\mathbf{k}, \omega) \left(\mathbf{j}(\mathbf{k}, \omega) - \frac{c^2 \mathbf{k}}{\omega} \rho(\mathbf{k}, \omega) \right). \quad (4.7)$$

This is equivalent to $A = D_0 j$ with the temporal Green function given by Eq. (3.56). Further using the inhomogeneous Maxwell equations to express the charge and current densities in terms of the electric and magnetic fields, we obtain an expression $\mathbf{A}[\mathbf{E}, \mathbf{B}]$, which we call the *canonical functional in the temporal gauge*:

$$\begin{aligned} \mathbf{A}(\mathbf{k}, \omega) &= -\varepsilon_0 \mathcal{D}_0(\mathbf{k}, \omega) \\ &\times \frac{1}{i\omega} \left(\omega^2 \mathbf{E}(\mathbf{k}, \omega) - c^2 \mathbf{k} (\mathbf{k} \cdot \mathbf{E}(\mathbf{k}, \omega)) + \omega c^2 \mathbf{k} \times \mathbf{B}(\mathbf{k}, \omega) \right). \end{aligned} \quad (4.8)$$

To simplify this expression, we define the *electric solution generator*

$$\overset{\leftrightarrow}{\mathbb{E}}(\mathbf{k}, \omega) = -\varepsilon_0 \omega^2 \mathcal{D}_0(\mathbf{k}, \omega) \left(\left(1 - \frac{c^2 |\mathbf{k}|^2}{\omega^2} \right) \overset{\leftrightarrow}{P}_L(\mathbf{k}) + \overset{\leftrightarrow}{P}_T(\mathbf{k}) \right) \quad (4.9)$$

$$= \overset{\leftrightarrow}{P}_L(\mathbf{k}) + \frac{\omega^2}{\omega^2 - c^2 |\mathbf{k}|^2} \overset{\leftrightarrow}{P}_T(\mathbf{k}), \quad (4.10)$$

and the *magnetic solution generator*

$$\overset{\leftrightarrow}{\mathbb{B}}(\mathbf{k}, \omega) = -\varepsilon_0 \omega^2 \mathcal{D}_0(\mathbf{k}, \omega) \left(\frac{c |\mathbf{k}|}{\omega} \overset{\leftrightarrow}{R}_T(\mathbf{k}) \right) \quad (4.11)$$

$$= \frac{\omega c |\mathbf{k}|}{\omega^2 - c^2 |\mathbf{k}|^2} \overset{\leftrightarrow}{R}_T(\mathbf{k}). \quad (4.12)$$

(The reason for this labeling will become clear at the end of this subsection.) These are dimensionless operators, which are given in components by

$$\mathbb{E}_{ij}(\mathbf{k}, \omega) = \frac{\omega^2 \delta_{ij} - c^2 k_i k_j}{\omega^2 - c^2 |\mathbf{k}|^2}, \quad (4.13)$$

$$\mathbb{B}_{ij}(\mathbf{k}, \omega) = \frac{\omega \epsilon_{ij\ell} c k_\ell}{\omega^2 - c^2 |\mathbf{k}|^2}. \quad (4.14)$$

In terms of the electric and magnetic solution generators, the canonical functional (4.8) can be written as

$$\mathbf{A}(\mathbf{k}, \omega) = \frac{1}{i\omega} \left(\overset{\leftrightarrow}{\mathbb{E}}(\mathbf{k}, \omega) \mathbf{E}(\mathbf{k}, \omega) + \overset{\leftrightarrow}{\mathbb{B}}(\mathbf{k}, \omega) c \mathbf{B}(\mathbf{k}, \omega) \right). \quad (4.15)$$

Equivalently, it is given in component form by

$$A_i(\mathbf{k}, \omega) = \frac{1}{i\omega} \frac{\omega^2 \delta_{ij} - c^2 k_i k_j}{\omega^2 - c^2 |\mathbf{k}|^2} E_j(\mathbf{k}, \omega) + \frac{1}{i\omega} \frac{\omega \epsilon_{ilj} c k_\ell}{\omega^2 - c^2 |\mathbf{k}|^2} c B_j(\mathbf{k}, \omega). \quad (4.16)$$

In particular, we read off the partial functional derivatives of the vector potential with respect to the electric and magnetic fields,

$$\frac{\delta A_i(\mathbf{k}, \omega)}{\delta E_j(\mathbf{k}', \omega')} = \frac{c}{i\omega} \frac{\omega^2 \delta_{ij} - c^2 k_i k_j}{\omega^2 - c^2 |\mathbf{k}|^2} \delta^3(\mathbf{k} - \mathbf{k}') \delta(\omega - \omega'), \quad (4.17)$$

$$\frac{1}{c} \frac{\delta A_i(\mathbf{k}, \omega)}{\delta B_j(\mathbf{k}', \omega')} = \frac{c}{i\omega} \frac{\omega \epsilon_{ilj} c k_\ell}{\omega^2 - c^2 |\mathbf{k}|^2} \delta^3(\mathbf{k} - \mathbf{k}') \delta(\omega - \omega'). \quad (4.18)$$

Our formalism is consistent in that the same functional $\mathbf{A}[\mathbf{E}, \mathbf{B}]$ is obtained if we first use the continuity equation to eliminate ρ in terms of \mathbf{j} in Eq. (4.7),

$$\mathbf{A}(\mathbf{k}, \omega) = \mathbb{D}_0(\mathbf{k}, \omega) \left(\mathbf{j}(\mathbf{k}, \omega) - \frac{c^2 \mathbf{k}(\mathbf{k} \cdot \mathbf{j}(\mathbf{k}, \omega))}{\omega^2} \right), \quad (4.19)$$

and then express the current in terms of the electric and magnetic fields by Ampère's law. The reason for this is that the continuity equation and Ampère's law imply

$$\rho(\mathbf{k}, \omega) = \frac{\mathbf{k} \cdot \mathbf{j}(\mathbf{k}, \omega)}{\omega} = \varepsilon_0 i \mathbf{k} \cdot \mathbf{E}(\mathbf{k}, \omega), \quad (4.20)$$

which is the same expression for ρ in terms of \mathbf{E} as given by Gauss's law.

We conclude this subsection with a remark about the electric and magnetic solution generators defined in Eqs. (4.9) and (4.11). Apart from the canonical functional, these operators also appear in the equations of motion for the electric and magnetic fields in terms of the sources: Inverting Eqs. (2.51)–(2.52) yields the expressions in Fourier space

$$\mathbf{E}(\mathbf{k}, \omega) = \mathbb{D}_0(\mathbf{k}, \omega) \left(-c^2 i \mathbf{k} \rho(\mathbf{k}, \omega) + i \omega \mathbf{j}(\mathbf{k}, \omega) \right), \quad (4.21)$$

$$\mathbf{B}(\mathbf{k}, \omega) = \mathbb{D}_0(\mathbf{k}, \omega) \left(i \mathbf{k} \times \mathbf{j}(\mathbf{k}, \omega) \right). \quad (4.22)$$

Using the continuity equation to eliminate ρ in terms of \mathbf{j} in the first equation, we find that these equations are equivalent to

$$\mathbf{E}(\mathbf{k}, \omega) = \frac{1}{\varepsilon_0} \frac{1}{i\omega} \overset{\leftrightarrow}{E}(\mathbf{k}, \omega) \mathbf{j}(\mathbf{k}, \omega), \quad (4.23)$$

$$c\mathbf{B}(\mathbf{k}, \omega) = \frac{1}{\varepsilon_0} \frac{1}{i\omega} \overset{\leftrightarrow}{B}(\mathbf{k}, \omega) \mathbf{j}(\mathbf{k}, \omega). \quad (4.24)$$

Thus, with the electric and magnetic solution generators one can formally solve the Maxwell equations in terms of the spatial current. The relevance of the above expressions will become clear in Sections 5.4 and 6.6.

4.2. Total functional derivatives

The expansion coefficients of \mathbf{E} and \mathbf{B} in the canonical functional, Eqs. (4.17)–(4.18), correspond to partial derivatives of the vector potential, because these are obtained by varying \mathbf{E} and \mathbf{B} independently. Physically, however, \mathbf{E} and \mathbf{B} are not independent of each other: by Faraday's law, the magnetic field is related to the transverse part of the electric field as

$$\mathbf{B} = \frac{\mathbf{k} \times \mathbf{E}}{\omega}, \quad (4.25)$$

and conversely, the electric field can be decomposed as

$$\mathbf{E} = \mathbf{E}_L + \mathbf{E}_T = \mathbf{E}_L - \omega \frac{\mathbf{k} \times \mathbf{B}}{|\mathbf{k}|^2}. \quad (4.26)$$

Therefore, it is also possible to express the vector potential \mathbf{A} exclusively in terms of \mathbf{E} , or in terms of \mathbf{E}_L and \mathbf{B} . To show this explicitly, we rewrite the canonical functional $\mathbf{A}[\mathbf{E}, \mathbf{B}]$ from Eq. (4.8) as

$$\begin{aligned} \mathbf{A}(\mathbf{k}, \omega) &= \frac{1}{i\omega} \mathbf{E}_L(\mathbf{k}, \omega) \\ &+ \frac{1}{i\omega} \frac{1}{\omega^2 - c^2|\mathbf{k}|^2} \left(\omega^2 \mathbf{E}_T(\mathbf{k}, \omega) + \omega c^2 \mathbf{k} \times \mathbf{B}(\mathbf{k}, \omega) \right). \end{aligned} \quad (4.27)$$

Eliminating \mathbf{B} in terms of \mathbf{E}_T or vice versa, we find

$$\mathbf{A}[\mathbf{E}] = \frac{1}{i\omega} \mathbf{E}_L + \frac{1}{i\omega} \mathbf{E}_T = \frac{1}{i\omega} \mathbf{E}, \quad (4.28)$$

$$\mathbf{A}[\mathbf{E}_L, \mathbf{B}] = \frac{1}{i\omega} \mathbf{E}_L + \frac{i\mathbf{k} \times \mathbf{B}}{|\mathbf{k}|^2}. \quad (4.29)$$

In matrix notation, using the transverse rotation operator defined in Section 2.1, we can write these equations equivalently as

$$\mathbf{A}(\mathbf{k}, \omega) = \frac{1}{i\omega} \mathbf{E}(\mathbf{k}, \omega), \quad (4.30)$$

$$\mathbf{A}(\mathbf{k}, \omega) = \frac{1}{i\omega} \mathbf{E}_L(\mathbf{k}, \omega) + \frac{1}{i\omega} \left(-\frac{\omega}{c|\mathbf{k}|} \right) \overset{\leftrightarrow}{R}_T(\mathbf{k}) c\mathbf{B}(\mathbf{k}, \omega). \quad (4.31)$$

Finally, we can write them in component form as

$$A_i(\mathbf{k}, \omega) = \frac{1}{i\omega} E_i(\mathbf{k}, \omega), \quad (4.32)$$

$$A_i(\mathbf{k}, \omega) = \frac{1}{i\omega} (E_L)_i(\mathbf{k}, \omega) + \frac{1}{i\omega} \left(\frac{\omega \epsilon_{i\ell j} c k_\ell}{-c^2 |\mathbf{k}|^2} \right) c B_j(\mathbf{k}, \omega). \quad (4.33)$$

These relations can also be obtained directly from Eqs. (4.4)–(4.5), which read in Fourier space $\mathbf{E} = i\omega \mathbf{A}$ and $\mathbf{B} = i\mathbf{k} \times \mathbf{A}$: Inverting the first equation yields immediately (4.28). Moreover, using the decomposition of \mathbf{A} into longitudinal and transverse parts,

$$\mathbf{A} = \frac{\mathbf{k}(\mathbf{k} \cdot \mathbf{A})}{|\mathbf{k}|^2} - \frac{\mathbf{k} \times (\mathbf{k} \times \mathbf{A})}{|\mathbf{k}|^2} = \frac{1}{i\omega} \frac{\mathbf{k}(\mathbf{k} \cdot \mathbf{E})}{|\mathbf{k}|^2} + \frac{i\mathbf{k} \times \mathbf{B}}{|\mathbf{k}|^2}, \quad (4.34)$$

and identifying the first term on the right hand side with $\mathbf{E}_L/i\omega$, we recover again (4.29). From this alternative derivation we conclude that Eqs. (4.28) and (4.29) are in fact generally valid and apply in particular to vacuum fields, although the canonical functional (4.8) had originally been constructed for retarded fields generated by sources. Vacuum fields have a conceptual relevance, because in some situations it is an appropriate idealization to consider the external perturbations as free electromagnetic waves (although strictly speaking these do not have sources and hence cannot be produced in the laboratory).

We now define the *total functional derivatives* of the vector potential with respect to the electric and magnetic fields as

$$\frac{dA_i}{dE_j} = \frac{\delta A_i}{\delta E_j} + \frac{\delta A_i}{\delta B_\ell} \frac{\delta B_\ell}{\delta E_j}, \quad (4.35)$$

$$\frac{dA_i}{dB_j} = \frac{\delta A_i}{\delta B_j} + \frac{\delta A_i}{\delta E_\ell} \frac{\delta E_\ell}{\delta B_j}. \quad (4.36)$$

Here, the functional derivatives of the magnetic field with respect to the electric field and vice versa are given by Eqs. (4.25)–(4.26) as

$$c \frac{\delta B_\ell(\mathbf{k}, \omega)}{\delta E_j(\mathbf{k}, \omega)} = \frac{c|\mathbf{k}|}{\omega} \epsilon_{\ell nj} \frac{k_n}{|\mathbf{k}|} \quad (4.37)$$

and

$$\frac{1}{c} \frac{\delta E_\ell(\mathbf{k}, \omega)}{\delta B_j(\mathbf{k}, \omega)} = \frac{1}{c} \frac{\delta (E_T)_\ell(\mathbf{k}, \omega)}{\delta B_j(\mathbf{k}, \omega)} = -\frac{\omega}{c|\mathbf{k}|} \epsilon_{\ell nj} \frac{k_n}{|\mathbf{k}|}. \quad (4.38)$$

The importance of the total functional derivatives lies in the fact that they lead to physical response functions, as we will argue in Section 6.1. They can be calculated from Eqs. (4.35)–(4.36) by using the formulae for the partial derivatives (4.17)–(4.18) derived in the previous subsection together with Eqs. (4.37)–(4.38). More easily, we can read them off directly from the representations (4.28)–(4.29) of the vector potential. We thus obtain the following formulae:

$$\frac{dA_i(\mathbf{k}, \omega)}{dE_j(\mathbf{k}', \omega')} = \frac{c}{i\omega} \delta_{ij} \delta^3(\mathbf{k} - \mathbf{k}') \delta(\omega - \omega'), \quad (4.39)$$

$$\frac{1}{c} \frac{dA_i(\mathbf{k}, \omega)}{dB_j(\mathbf{k}', \omega')} = \frac{c}{i\omega} \left(\frac{\omega \epsilon_{ilj} c k_\ell}{-c^2 |\mathbf{k}|^2} \right) \delta^3(\mathbf{k} - \mathbf{k}') \delta(\omega - \omega'). \quad (4.40)$$

In real space, they can be written as

$$\frac{dA_i(\mathbf{x}, t)}{dE_j(\mathbf{x}', t')} = -\frac{1}{c} \delta_{ij} \delta^3(\mathbf{x} - \mathbf{x}') \theta(t - t'), \quad (4.41)$$

$$\frac{1}{c} \frac{dA_i(\mathbf{x}, t)}{dB_j(\mathbf{x}', t')} = \frac{1}{4\pi c^2} \epsilon_{ilj} \frac{\partial}{\partial x^\ell} \frac{\delta(t - t')}{|\mathbf{x} - \mathbf{x}'|}, \quad (4.42)$$

where $\theta(t - t')$ denotes the Heaviside step function.

4.3. Zero frequency limit

We now come to a problem which we have ignored so far, namely the divergence of many formal expressions in the limit $\omega \rightarrow 0$. While the zero frequency limit can be performed without any problems for quantities like the scalar Green function $\mathbb{D}_0(\mathbf{k}, \omega)$ (as long as $\mathbf{k} \neq 0$), the same does not apply to the operator $1/i\omega$ which connects, for example, the vector potential $\mathbf{A}(\mathbf{k}, \omega)$ to the electric field $\mathbf{E}(\mathbf{k}, \omega)$ in Eq. (4.30). When it comes to the

limit $\omega \rightarrow 0$, these expressions have to be regularized. As this can be done in different ways, we first consider the problem in real space, where the vector potential can be defined in terms of the electric and magnetic fields by an initial-value problem: it satisfies for $t > t_0$ the equations

$$-\partial_t \mathbf{A}(\mathbf{x}, t) = \mathbf{E}(\mathbf{x}, t), \quad (4.43)$$

$$\nabla \times \mathbf{A}(\mathbf{x}, t) = \mathbf{B}(\mathbf{x}, t), \quad (4.44)$$

and is subject to the initial condition

$$\mathbf{A}(\mathbf{x}, t_0) = \mathbf{A}_0(\mathbf{x}). \quad (4.45)$$

The latter cannot be chosen arbitrarily, but is constrained by

$$\nabla \times \mathbf{A}_0(\mathbf{x}) = \mathbf{B}_0(\mathbf{x}) \equiv \mathbf{B}(\mathbf{x}, t_0). \quad (4.46)$$

We may choose \mathbf{A}_0 purely transverse, such that it is given explicitly in terms of \mathbf{B}_0 by

$$\mathbf{A}_0(\mathbf{x}) = \frac{1}{4\pi} \nabla \times \int d^3 \mathbf{x}' \frac{\mathbf{B}_0(\mathbf{x}')}{|\mathbf{x} - \mathbf{x}'|}. \quad (4.47)$$

The unique solution to the initial value problem (4.43)–(4.46) is then given by

$$\mathbf{A}(\mathbf{x}, t) = - \int_{t_0}^t dt' \mathbf{E}(\mathbf{x}, t') + \mathbf{A}_0(\mathbf{x}). \quad (4.48)$$

While Eq. (4.43) is obviously fulfilled, Eq. (4.44) follows by Faraday's law,

$$(\nabla \times \mathbf{A})(\mathbf{x}, t) = - \int_{t_0}^t dt' (\nabla \times \mathbf{E})(\mathbf{x}, t') + \nabla \times \mathbf{A}_0(\mathbf{x}) \quad (4.49)$$

$$= \int_{t_0}^t dt' \partial_{t'} \mathbf{B}(\mathbf{x}, t') + \mathbf{B}(\mathbf{x}, t_0) \quad (4.50)$$

$$= \mathbf{B}(\mathbf{x}, t). \quad (4.51)$$

This discussion shows that in general, the vector potential \mathbf{A} cannot be expressed entirely in terms of the electric field \mathbf{E} . Instead, it is a functional of the electric field *and the initial magnetic field* \mathbf{B}_0 . For a typical experimental setup where the external and induced quantities vanish before some initial switching-on, this does not pose any problem, because if we choose t_0

sufficiently early, the initial fields can be set to zero (see the discussion in Section 3.2). Unfortunately, however, this assumption precludes the possibility of static fields which have the same value at all times. Strictly speaking, such static fields cannot be produced in the laboratory, but for many situations they are a suitable idealization. It is therefore desirable to allow more generally for external and induced fields which do not vanish before any initial time t_0 . We are thus led to consider the more general case of fields with a nonvanishing *static contribution*

$$\mathbf{B}_0(\mathbf{x}) = \lim_{t_0 \rightarrow -\infty} \mathbf{B}(\mathbf{x}, t_0), \quad (4.52)$$

and analogously for the vector potential. The solution (4.48) can be written in the limit $t_0 \rightarrow -\infty$ as

$$\mathbf{A}(\mathbf{x}, t) = - \int_{-\infty}^{\infty} dt' \theta(t - t') \mathbf{E}(\mathbf{x}, t') + \mathbf{A}_0(\mathbf{x}). \quad (4.53)$$

This equation shows that for the initial value problem at hand, the negative Heaviside step function plays the rôle of the retarded Green function, while the propagator is given by the identity operator (cf. Section 3.1). Using that

$$\theta(t - t') = i \int \frac{d\omega}{2\pi} \frac{e^{-i\omega(t-t')}}{\omega + i\eta}, \quad (4.54)$$

we can write this solution in the momentum and frequency domain as

$$\mathbf{A}(\mathbf{k}, \omega) = \frac{1}{i(\omega + i\eta)} \mathbf{E}(\mathbf{k}, \omega) + \frac{i\mathbf{k} \times \mathbf{B}_0(\mathbf{k})}{|\mathbf{k}|^2} \delta(\omega). \quad (4.55)$$

In fact, this is a distributional solution to the equation

$$i\omega \mathbf{A}(\mathbf{k}, \omega) = \mathbf{E}(\mathbf{k}, \omega). \quad (4.56)$$

We conclude that in order to incorporate external magnetic fields with a static contribution, Eq. (4.30) should be replaced by Eq. (4.55). By contrast, if we consider fields produced in the laboratory after some initial switching-on, these fields vanish in the limit $t \rightarrow -\infty$, and hence we recover precisely our original formula (4.30) provided we regularize it by the prescription $\omega \mapsto \omega + i\eta$.

Next, let us derive also the functional $\mathbf{A}[\mathbf{E}_L, \mathbf{B}]$ allowing for a static contribution to the magnetic field. As indicated by the Dirac delta distribution in Eq. (4.55), for fields which do not vanish in the limit $t \rightarrow -\infty$, the Fourier transform becomes in general singular. Since formal manipulations of singular Fourier transforms may easily lead to nonsensical results, we have to proceed more carefully. We therefore avoid the frequency domain and instead start again from Eq. (4.48) in the time domain (where we let $t_0 \rightarrow -\infty$). We decompose the electric field under the integral into its longitudinal and transverse parts, and write the latter as

$$\mathbf{E}_T(\mathbf{x}, t) = \frac{1}{4\pi} \nabla \times \int d^3\mathbf{x}' \frac{-\partial_t \mathbf{B}(\mathbf{x}', t)}{|\mathbf{x} - \mathbf{x}'|}, \quad (4.57)$$

which is equivalent to Faraday's law. Plugging this into Eq. (4.48) and using Eq. (4.47), we observe that the two contributions from $\mathbf{B}_0(\mathbf{x})$ cancel, and we arrive at

$$\mathbf{A}(\mathbf{x}, t) = - \int_{-\infty}^t dt' \mathbf{E}_L(\mathbf{x}, t') + \frac{1}{4\pi} \nabla \times \int d^3\mathbf{x}' \frac{\mathbf{B}(\mathbf{x}', t)}{|\mathbf{x} - \mathbf{x}'|}. \quad (4.58)$$

In Fourier space, we can now rewrite this equation as

$$\mathbf{A}(\mathbf{k}, \omega) = \frac{1}{i(\omega + i\eta)} \mathbf{E}_L(\mathbf{k}, \omega) + \frac{i\mathbf{k} \times \mathbf{B}(\mathbf{k}, \omega)}{|\mathbf{k}|^2}. \quad (4.59)$$

This expression coincides precisely with our original formula (4.29) under the regularization prescription $\omega \mapsto \omega + i\eta$. Hence, in contrast to Eq. (4.30), this equation remains valid even in the presence of static magnetic fields.

In summary, we have introduced three ways to represent the vector potential in terms of the electric and magnetic fields:

$$\mathbf{A}[\mathbf{E}, \mathbf{B}] = \frac{\delta \mathbf{A}}{\delta \mathbf{E}} \mathbf{E} + \frac{\delta \mathbf{A}}{\delta \mathbf{B}} \mathbf{B}, \quad (4.60)$$

$$\mathbf{A}[\mathbf{E}, \mathbf{B}_0] = \frac{d\mathbf{A}}{d\mathbf{E}} \mathbf{E} + \frac{d\mathbf{A}}{d\mathbf{B}} \mathbf{B}_0, \quad (4.61)$$

$$\mathbf{A}[\mathbf{E}_L, \mathbf{B}] = \frac{d\mathbf{A}}{d\mathbf{E}} \mathbf{E}_L + \frac{d\mathbf{A}}{d\mathbf{B}} \mathbf{B}, \quad (4.62)$$

Here, we use the notation for 3×3 matrices

$$\left(\frac{\delta \mathbf{A}}{\delta \mathbf{E}} \right)_{ij} = \frac{\delta A_i}{\delta E_j}, \quad (4.63)$$

and $\mathbf{B}_0 \equiv \mathbf{B}_0(\mathbf{k}, \omega) = \mathbf{B}_0(\mathbf{k})\delta(\omega)$ denotes the static contribution to the magnetic field. The partial and the total functional derivatives appearing in the above expansions are given explicitly in the temporal gauge by Eqs. (4.17)–(4.18) and by Eqs. (4.39)–(4.40), respectively. In the following section, we will use these expansions to derive electromagnetic material properties from the current response to an external vector potential. Moreover, we will show in Section 6.6 that the three different representations of the vector potential lead to three different but equivalent expansions of the induced fields in terms of the external electric and magnetic fields.

5. Electrodynamics of materials

5.1. Fundamental response functions

In a general experimental setup, the electromagnetic response of a material probe is determined under an externally applied electromagnetic perturbation. For the whole system comprising the probe and the external perturbation, this means that all electromagnetic quantities (such as fields, charges and currents) are split into *internal* and *external* contributions. Thinking of the external perturbation as inducing a redistribution of charges and currents in the sample, a natural starting point for the theoretical description is the functional (see, e.g., [88, 89])

$$j_{\text{int}}^\mu = j_{\text{int}}^\mu[A_{\text{ext}}^\nu], \quad (5.1)$$

where j_{int}^μ is the four-current of the medium and A_{ext}^ν the four-potential of the perturbation. It is inherent in this definition that on the most fundamental level, the induced currents and charges are not functionals of the external electric and magnetic fields but of the electromagnetic potentials. The main arguments in favor of this viewpoint are: (i) In general, the electric and magnetic fields are not independent of each other, hence the functional $j_{\text{int}}^\mu[\mathbf{E}_{\text{ext}}, \mathbf{B}_{\text{ext}}]$ is a priori not well-defined, (ii) the functional (5.1) connects two Lorentz four-vectors and therefore leads to a manifestly Lorentz covariant description of the electromagnetic response, and (iii) it is the external four-potential which couples to the microscopic degrees of freedom of the system, namely by the minimal coupling prescription $\partial^\mu \mapsto \partial^\mu + ieA_{\text{ext}}^\mu/\hbar$ in the fundamental Hamiltonian of the material.

In general, the functional dependence (5.1) may be complicated, depend on past history (hysteresis), be nonlinear, etc. [3]. Here we only assume that

it is differentiable, hence up to first order the Taylor series expansion reads

$$j_{\text{int}}^\mu(x) = j_{\text{int},0}^\mu(x) + \int d^4x' \frac{\delta j_{\text{int}}^\mu(x)}{\delta A_{\text{ext}}^\nu(x')} (A_{\text{ext}}^\nu(x') - A_{\text{ext},0}^\nu(x')). \quad (5.2)$$

In this equation, the functional derivative is to be evaluated at the reference potential $A_{\text{ext},0}^\nu$. The difference between internal currents in the presence and in the absence of the external perturbation is called the *induced four-current*,

$$j_{\text{ind}}^\mu = j_{\text{int}}^\mu - j_{\text{int},0}^\mu. \quad (5.3)$$

The above expansion is typically performed around $A_{\text{ext},0}^\nu \equiv 0$, but it is also possible to consider small perturbations around a finite reference potential. (In this way one can describe, for example, the current response to an applied voltage in the presence of a constant external magnetic field, i.e. the Hall conductivity [90].) From the functional point of view, linear response theory restricts attention to the first order terms, i.e. to the calculation of the functional derivatives

$$\chi^\mu{}_\nu(x, x') = \frac{\delta j_{\text{ind}}^\mu(x)}{\delta A_{\text{ext}}^\nu(x')}, \quad (5.4)$$

which by the discussion in Section 3.2 are causal (i.e. retarded) response functions. We will refer to this 4×4 tensor as the *fundamental response tensor*, and to its components as the *fundamental response functions*. We do not presume that the system under consideration actually behaves linearly, but instead we will make generally valid statements about the first order derivatives of induced quantities with respect to external perturbations.

The following constraints on $\chi^\mu{}_\nu$ are required by the continuity equation and by the gauge invariance of the induced current, respectively [88, 91]:

$$\partial_\mu \chi^\mu{}_\nu(x, x') = 0, \quad (5.5)$$

$$\partial^\nu \chi^\mu{}_\nu(x, x') = 0. \quad (5.6)$$

Assuming homogeneity in time, these constraints are equivalent to

$$\frac{1}{c} \frac{\partial}{\partial t} \chi^0{}_\nu(\mathbf{x}, \mathbf{x}'; t - t') + \sum_i \frac{\partial}{\partial x^i} \chi^i{}_\nu(\mathbf{x}, \mathbf{x}'; t - t') = 0, \quad (5.7)$$

$$-\frac{1}{c} \frac{\partial}{\partial t'} \chi^\mu{}_0(\mathbf{x}, \mathbf{x}'; t - t') + \sum_j \frac{\partial}{\partial x'^j} \chi^\mu{}_j(\mathbf{x}, \mathbf{x}'; t - t') = 0. \quad (5.8)$$

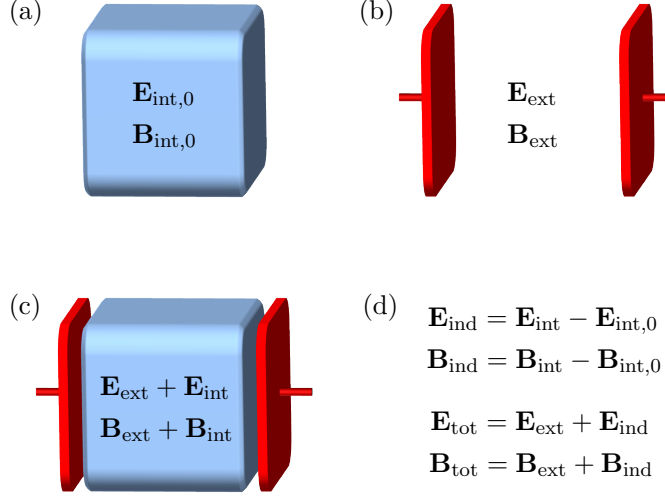


Figure 2: Definition of external, internal, induced and total fields (cf. [1, 56]). (a) ($\mathbf{E}_{\text{int},0}$, $\mathbf{B}_{\text{int},0}$) are the fields produced by the internal sources, i.e., the charges and currents constituting the material sample (without the external perturbation). (b) The *external fields* are produced by the external sources in the absence of the medium. (c) In the presence of the external sources, the fields produced by the (redistributed) internal sources are a superposition of the external and the *internal fields*. (d) Subtracting from the latter the fields ($\mathbf{E}_{\text{int},0}$, $\mathbf{B}_{\text{int},0}$) yields the *induced fields*. Finally, the *total fields* are defined as the sum of the external and the induced fields.

Therefore, at most 9 of the 16 fundamental response functions are independent of each other (cf. [10, 28, 29]). Explicitly, we can express all components χ^μ_ν in terms of the spatial components $\chi^i_j \equiv \chi_{ij}$ as

$$\chi^0_j(\mathbf{x}, \mathbf{x}'; \omega) = \frac{c}{i\omega} \sum_i \frac{\partial}{\partial x^i} \chi_{ij}(\mathbf{x}, \mathbf{x}'; \omega), \quad (5.9)$$

$$\chi^i_0(\mathbf{x}, \mathbf{x}'; \omega) = \frac{c}{i\omega} \sum_j \frac{\partial}{\partial x'^j} \chi_{ij}(\mathbf{x}, \mathbf{x}'; \omega), \quad (5.10)$$

$$\chi^0_0(\mathbf{x}, \mathbf{x}'; \omega) = -\frac{c^2}{\omega^2} \sum_{i,j} \frac{\partial}{\partial x^i} \frac{\partial}{\partial x'^j} \chi_{ij}(\mathbf{x}, \mathbf{x}'; \omega). \quad (5.11)$$

In Fourier space, the fundamental response tensor can therefore be written

compactly as

$$\chi_{\nu}^{\mu}(\mathbf{k}, \mathbf{k}', \omega) = \begin{pmatrix} -\frac{c^2}{\omega^2} \mathbf{k}^T \overleftrightarrow{\chi} \mathbf{k}' & \frac{c}{\omega} \mathbf{k}^T \overleftrightarrow{\chi} \\ -\frac{c}{\omega} \overleftrightarrow{\chi} \mathbf{k}' & \overleftrightarrow{\chi} \end{pmatrix}. \quad (5.12)$$

Apart from reducing the number of independent response functions, focusing on the fundamental response functions in the first place bears several advantages: They constitute a second-rank Lorentz tensor and thus yield a relativistic description of the electromagnetic response. This is especially relevant for the study of moving media [92–94] (see also [95] for a discussion of Ohm’s law). The relativistic transformation law for the fundamental response functions reads explicitly

$$\chi_{\nu}^{\mu}(x, y) = \Lambda^{\mu}_{\mu'} \Lambda_{\nu'}^{\nu'} \chi^{\mu'}_{\nu'}(\Lambda^{-1}x, \Lambda^{-1}y), \quad (5.13)$$

with Lorentz transformations $\Lambda \in \text{O}(3, 1)$. Furthermore, the minimal coupling prescription implies that the fundamental response functions can be calculated directly in the Kubo formalism (see, e.g., [11, Eqs. (2.154)–(2.155)] and [88, 96]). However, the crucial question is whether one can derive closed expressions for the usual electromagnetic response functions (such as the conductivity, the dielectric tensor and the magnetic susceptibility) in terms of the fundamental response functions. This question will be addressed in Sections 5.3 and 6.

5.2. Connection to Schwinger–Dyson and Hedin equations

As described in the previous subsection, the Functional Approach to electrodynamics of materials is based on the splitting of all electromagnetic quantities into external and internal (or induced) contributions. Each of these subsystems can be described equivalently by its charges and currents $j^{\mu} = (c\rho, \mathbf{j})$, by its gauge potential $A^{\mu} = (\varphi/c, \mathbf{A})$, or by its electric and magnetic fields $\{\mathbf{E}, \mathbf{B}\}$. The mutual functional dependencies of these different field quantities have been derived explicitly in the first part of this paper. In particular, the *induced electromagnetic fields* are uniquely determined as functionals of the induced charges and currents, which were defined in the previous subsection (see Section 3.2). Schematically, the external, internal, induced and total fields are shown in Fig. 2.

Now given the functional dependence of the induced four-current on the external four-potential, it follows that all other electromagnetic response

functions (specifying, for example, the induced current in terms of the external electric field, or in terms of the external charges and currents) can be calculated. Moreover, Eq. (5.4) defines only one of a number of equivalent response tensors: instead of describing the response of the system in terms of the induced quantities, one may as well characterize it by the functional dependence of the total on the external, or even the induced on the total field quantities. Each of these possibilities gives rise to an equivalent set of response functions, all of which are mutually related by functional chain rules. As an example, we consider the converse point of view to Eq. (5.4), namely the dependence of the total four-potential on the external four-current. We define the *full tensorial Green function* as

$$D^\mu{}_\nu(x, x') = \frac{\delta A_{\text{tot}}^\mu(x)}{\delta j_{\text{ext}}^\nu(x')} . \quad (5.14)$$

A straightforward application of the functional chain rule in the form

$$\frac{\delta A_{\text{tot}}}{\delta j_{\text{ext}}} = \frac{\delta A_{\text{ext}}}{\delta j_{\text{ext}}} + \frac{\delta A_{\text{ind}}}{\delta j_{\text{ind}}} \frac{\delta j_{\text{ind}}}{\delta A_{\text{ext}}} \frac{\delta A_{\text{ext}}}{\delta j_{\text{ext}}} \quad (5.15)$$

leads to the equation

$$D = D_0 + D_0 \chi D_0 . \quad (5.16)$$

This relates D to the fundamental response tensor χ via the free tensorial Green function D_0 , which was defined in Section 3.3. Here we have used that $A = D_0 j$ implies in particular $D_0 = \delta A / \delta j$ (cf. [97, p. 21]). Note that the formal products refer to the natural multiplication in the linear space of 4×4 tensorial integral kernels.

Furthermore, we introduce the *proper response functions* $\tilde{\chi}$ which specify the response of the induced four-current to the total (instead of the external) four-potential,

$$\tilde{\chi}^\mu{}_\nu(x, x') = \frac{\delta j_{\text{ind}}^\mu(x)}{\delta A_{\text{tot}}^\nu(x')} . \quad (5.17)$$

Again, the functional chain rule

$$\frac{\delta j_{\text{ind}}}{\delta A_{\text{ext}}} = \frac{\delta j_{\text{ind}}}{\delta A_{\text{tot}}} \frac{\delta A_{\text{tot}}}{\delta A_{\text{ext}}} = \frac{\delta j_{\text{ind}}}{\delta A_{\text{tot}}} + \frac{\delta j_{\text{ind}}}{\delta A_{\text{tot}}} \frac{\delta A_{\text{ind}}}{\delta j_{\text{ind}}} \frac{\delta j_{\text{ind}}}{\delta A_{\text{ext}}} \quad (5.18)$$

shows that these quantities are related to the fundamental response functions through

$$\chi = \tilde{\chi} + \tilde{\chi} D_0 \chi . \quad (5.19)$$

In terms of these proper response functions, the full tensorial Green function D satisfies

$$D = D_0 + D_0 \tilde{\chi} D, \quad (5.20)$$

as follows again by a functional chain rule. This last equation has a Dyson-like structure and formally coincides with the Schwinger–Dyson equation for the full tensorial Green function in the sense of quantum electrodynamics [82, 98], provided one identifies $\tilde{\chi}$ with the irreducible photon self-energy. In fact, the photon self-energy is given by the current-current correlation function evaluated in the vacuum [99], which corresponds to the Kubo formula for the fundamental response function χ . This shows the literal analogy between solid state physics and quantum electrodynamics, where the vacuum plays the rôle of the polarizable material. Moreover, our analysis provides via Eq. (5.14) a classical interpretation of the full tensorial Green function: just as D_0 yields the electromagnetic four-potential in terms of its own sources, D yields the total four-potential in terms of the external four-current, i.e. to linear order,

$$A_{\text{tot}}^\mu = D^\mu{}_\nu j_{\text{ext}}^\nu. \quad (5.21)$$

We note, however, that in quantum electrodynamics one usually works with time-ordered instead of retarded response functions.

A non-relativistic version of the Schwinger–Dyson equations is known in electronic structure theory as *Hedin’s equations* [100, 101]. These are among the most important first-principles techniques used nowadays for describing single-electron excitations in real materials (see [102, 103], or [104] for a recent discussion in the context of Green function theory). They can be obtained from the relativistic Schwinger–Dyson equations by replacing the electronic Green function of the Dirac equation by the Green function of the Schrödinger or the Pauli equation and approximating the full tensorial Green function in the Coulomb gauge by its 00-component. Indeed, by keeping in Eq. (5.20) only the Coulomb potential (cf. Eq. (3.52))

$$v = c^2 (D_0)^0{}_0 \quad (5.22)$$

and the proper density response function

$$\tilde{\chi} = \frac{1}{c^2} \tilde{\chi}^0{}_0, \quad (5.23)$$

we obtain the approximate relation for $w = c^2 D^0{}_0$:

$$w = v + v \tilde{\chi} w. \quad (5.24)$$

This is precisely Hedin's equation for the *screened potential* w , where $\tilde{\chi}$ is known in electronic structure theory as the (*irreducible*) *polarizability* [65, 105].

5.3. Material properties from fundamental response functions

In electrodynamics of materials, the response of the polarization \mathbf{P} and the magnetization \mathbf{M} to the external electric and magnetic fields \mathbf{E}_{ext} and \mathbf{B}_{ext} is especially relevant. Hence, it is desirable to derive these material properties directly from the fundamental response functions. Here, we are facing two basic questions: (i) How is the response of \mathbf{P} and \mathbf{M} related to the induced four-current j_{ind}^μ , and (ii) how do we express the response to \mathbf{E}_{ext} and \mathbf{B}_{ext} in terms of the response to the external four-potential A_{ext}^ν . The first problem is solved under the identifications (1.9)–(1.14): The Maxwell equations for the induced fields (1.15)–(1.18) imply the equations of motion

$$\square \mathbf{P}(\mathbf{x}, t) = \nabla \rho_{\text{ind}}(\mathbf{x}, t) + \frac{1}{c^2} \frac{\partial}{\partial t} \mathbf{j}_{\text{ind}}(\mathbf{x}, t), \quad (5.25)$$

$$\square \mathbf{M}(\mathbf{x}, t) = \nabla \times \mathbf{j}_{\text{ind}}(\mathbf{x}, t). \quad (5.26)$$

By using the retarded Green function for the scalar wave equation, Eq. (3.18), we thus obtain the induced fields in terms of the induced currents. As for the second problem, it is natural to use the functional chain rules

$$\frac{\delta j_{\text{ind}}^\mu(x)}{\delta E_{\text{ext}}^\ell(x')} = \int d^4 y \frac{\delta j_{\text{ind}}^\mu(x)}{\delta A_{\text{ext}}^\alpha(y)} \frac{\delta A_{\text{ext}}^\alpha(y)}{\delta E_{\text{ext}}^\ell(x')}, \quad (5.27)$$

$$\frac{\delta j_{\text{ind}}^\mu(x)}{\delta B_{\text{ext}}^\ell(x')} = \int d^4 y \frac{\delta j_{\text{ind}}^\mu(x)}{\delta A_{\text{ext}}^\alpha(y)} \frac{\delta A_{\text{ext}}^\alpha(y)}{\delta B_{\text{ext}}^\ell(x')}, \quad (5.28)$$

and to evaluate these formulae using the functional defined in Section 4.1,

$$A^\mu[\mathbf{E}, \mathbf{B}] = \frac{1}{2\mu_0} (\partial_\lambda (D_0)^\mu{}_\nu - \partial_\nu (D_0)^\mu{}_\lambda) F^{\nu\lambda} + (\partial^\mu f)[\mathbf{E}, \mathbf{B}], \quad (5.29)$$

where $F^{\nu\lambda}$ is expressed explicitly in terms of \mathbf{E} and \mathbf{B} by Eq. (2.41). However, as argued before, this functional is not uniquely determined for two reasons: (i) It contains an arbitrary pure gauge $\partial^\mu f$ (where the function f may itself depend on \mathbf{E} and \mathbf{B}), and (ii) there are infinitely many choices for the tensorial Green function D_0 . We now have to show that despite this ambiguity of the functional $A^\mu[\mathbf{E}, \mathbf{B}]$, the electromagnetic response functions

(5.27)–(5.28) are well-defined. In order to do this, we first note that no matter how we choose $f[\mathbf{E}, \mathbf{B}]$ and D_0 , the resulting four-potential produces the same electromagnetic fields, namely for $A^\mu = (\varphi/c, \mathbf{A})$:

$$(-\nabla\varphi - \partial_t \mathbf{A})[\mathbf{E}, \mathbf{B}] = \mathbf{E}, \quad (5.30)$$

$$(\nabla \times \mathbf{A})[\mathbf{E}, \mathbf{B}] = \mathbf{B}. \quad (5.31)$$

Since any two four-potentials A^μ and A'^μ which produce the same electromagnetic fields differ by a pure gauge,

$$A^\mu[\mathbf{E}, \mathbf{B}] = A'^\mu[\mathbf{E}, \mathbf{B}] + (\partial^\mu \tilde{f})[\mathbf{E}, \mathbf{B}], \quad (5.32)$$

the arbitrariness in the choice of D_0 can be absorbed in the gauge freedom. Hence, it only remains to show that Eqs. (5.27)–(5.28) yield gauge-independent results. For this purpose, it suffices to convince oneself that for an arbitrary function $\tilde{f}[\mathbf{E}, \mathbf{B}]$ the following equation holds:

$$\int d^4y \chi^\mu{}_\alpha(x, y) \frac{\delta(\partial^\alpha \tilde{f})(y)}{\delta E_{\text{ext}}^\ell(x')} = 0 \quad (5.33)$$

(and the analogous equation for the magnetic field). Using that the functional derivative commutes with the partial derivative and performing a partial integration, this condition follows directly from the constraint (5.6) on the fundamental response functions.

5.4. Field strength response tensor

We now derive an explicit expression for the partial functional derivatives of the induced fields with respect to the external fields in terms of the fundamental response tensor. These partial derivatives will be related to the constitutive tensor of bi-anisotropic materials at the end of this subsection. We proceed in a manifestly relativistic framework: In fact, the response functions relating induced to external fields can be grouped into the *field strength response tensor*

$$\chi^{\mu\nu}{}_{\alpha\beta}(x, x') = \frac{\delta F_{\text{ind}}^{\mu\nu}(x)}{\delta F_{\text{ext}}^{\alpha\beta}(x')}. \quad (5.34)$$

By a functional chain rule, we can express this fourth rank tensor through the fundamental response tensor as

$$\chi^{\mu\nu}{}_{\alpha\beta}(x, x') = \int d^4y \int d^4y' \frac{\delta F_{\text{ind}}^{\mu\nu}(x)}{\delta j_{\text{ind}}^\lambda(y)} \chi^\lambda{}_\rho(y, y') \frac{\delta A_{\text{ext}}^\rho(y')}{\delta F_{\text{ext}}^{\alpha\beta}(x')}. \quad (5.35)$$

We calculate the functional derivatives in the integrand as described in the previous subsection: From Eq. (3.17), we first obtain

$$\frac{\delta F^{\mu\nu}(x)}{\delta j^\lambda(y)} = (\partial^\mu \mathbb{D}_0)(x-y) \eta^\nu{}_\lambda - (\partial^\nu \mathbb{D}_0)(x-y) \eta^\mu{}_\lambda. \quad (5.36)$$

On the other hand, Eq. (4.2) yields

$$\frac{\delta A^\rho(y')}{\delta F^{\alpha\beta}(x')} = \frac{1}{2\mu_0} (\partial'_\beta (D_0)^\rho{}_\alpha(y'-x') - \partial'_\alpha (D_0)^\rho{}_\beta(y'-x')), \quad (5.37)$$

where the partial derivatives act on the y' variables. As shown above, the result of Eq. (5.35) does not depend on the choice of the Green function D_0 in the functional $A^\rho[F^{\alpha\beta}]$, and by choosing the Feynman Green function (3.51) we obtain the simpler expression

$$\frac{\delta A^\rho(y')}{\delta F^{\alpha\beta}(x')} = \frac{1}{2\mu_0} (\partial'_\beta \mathbb{D}_0(y'-x') \eta^\rho{}_\alpha - \partial'_\alpha \mathbb{D}_0(y'-x') \eta^\rho{}_\beta). \quad (5.38)$$

Inserting Eqs. (5.36) and (5.38) into Eq. (5.35) and performing partial integrations with respect to the y and y' variables, we arrive at the formula

$$\begin{aligned} \chi^{\mu\nu}{}_{\alpha\beta}(x, x') &= \frac{1}{2\mu_0} \int d^4y \int d^4y' \mathbb{D}_0(x-y) \\ &\times \left(\partial^\mu \partial'_\alpha \chi^\nu{}_\beta(y, y') - \partial^\nu \partial'_\alpha \chi^\mu{}_\beta(y, y') \right. \\ &\quad \left. - \partial^\mu \partial'_\beta \chi^\nu{}_\alpha(y, y') + \partial^\nu \partial'_\beta \chi^\mu{}_\alpha(y, y') \right) \mathbb{D}_0(y'-x'). \end{aligned} \quad (5.39)$$

Equivalently, we can write this in momentum space as

$$\begin{aligned} \chi^{\mu\nu}{}_{\alpha\beta}(k, k') &= \frac{1}{2\mu_0} \mathbb{D}_0(k) \left(k^\mu \chi^\nu{}_\beta(k, k') k'_\alpha - k^\nu \chi^\mu{}_\beta(k, k') k'_\alpha \right. \\ &\quad \left. - k^\mu \chi^\nu{}_\alpha(k, k') k'_\beta + k^\nu \chi^\mu{}_\alpha(k, k') k'_\beta \right) \mathbb{D}_0(k'). \end{aligned} \quad (5.40)$$

This general, manifestly Lorentz covariant formula allows for the calculation of all partial derivatives of the induced fields with respect to the external fields in terms of the fundamental response functions. It fully takes into account the inhomogeneity, anisotropy and magnetoelectric coupling of the material

as well as relativistic retardation effects, and it establishes the connection to the experiment through the linear expansion

$$F_{\text{ind}}^{\mu\nu}(k) = \int d^4k' \chi^{\mu\nu}_{\alpha\beta}(k, k') F_{\text{ext}}^{\alpha\beta}(k'). \quad (5.41)$$

Spelling out the electric and magnetic field components of the (induced and external) field strength tensors, we obtain the equivalent expansions

$$\mathbf{E}_{\text{ind}}(k) = \int d^4k' \overset{\leftrightarrow}{\chi}_{EE}^{\text{p}}(k, k') \mathbf{E}_{\text{ext}}(k') + \int d^4k' \overset{\leftrightarrow}{\chi}_{EB}^{\text{p}}(k, k') c\mathbf{B}_{\text{ext}}(k'), \quad (5.42)$$

$$c\mathbf{B}_{\text{ind}}(k) = \int d^4k' \overset{\leftrightarrow}{\chi}_{BE}^{\text{p}}(k, k') \mathbf{E}_{\text{ext}}(k') + \int d^4k' \overset{\leftrightarrow}{\chi}_{BB}^{\text{p}}(k, k') c\mathbf{B}_{\text{ext}}(k'). \quad (5.43)$$

Here, the coefficient functions are given in terms of the field strength response tensor by

$$\frac{\delta E_{\text{ind}}^i(k)}{\delta E_{\text{ext}}^j(k')} \equiv (\chi_{EE}^{\text{p}})^i_j(k, k') = 2\chi^{0i}_{0j}(k, k'), \quad (5.44)$$

$$\frac{1}{c} \frac{\delta E_{\text{ind}}^i(k)}{\delta B_{\text{ext}}^j(k')} \equiv (\chi_{EB}^{\text{p}})^i_j(k, k') = \epsilon_{j\ell n} \chi^{0i}_{\ell n}(k, k'), \quad (5.45)$$

$$c \frac{\delta B_{\text{ind}}^i(k)}{\delta E_{\text{ext}}^j(k')} \equiv (\chi_{BE}^{\text{p}})^i_j(k, k') = \epsilon_{ikm} \chi^{km}_{0j}(k, k'), \quad (5.46)$$

$$\frac{\delta B_{\text{ind}}^i(k)}{\delta B_{\text{ext}}^j(k')} \equiv (\chi_{BB}^{\text{p}})^i_j(k, k') = \frac{1}{2} \epsilon_{ikm} \epsilon_{j\ell n} \chi^{km}_{\ell n}(k, k'). \quad (5.47)$$

The superscript p indicates *partial* functional derivatives, meaning that the external electric and magnetic fields are varied independently of each other. By contrast, any physical response function should be identified with a *total* functional derivative, as we will argue in Section 6.1.

By our formula (5.40) we have derived a closed expression for the field strength response tensor in terms of the fundamental response tensor. As shown in Section 5.1, the fundamental response tensor has only 9 independent components. Therefore, it is also possible to express all components of the field strength response tensor in terms of only 9 independent current response functions. We will now derive explicit expressions for the 36 partial derivatives (5.44)–(5.47) in terms of the 9 spatial components χ_{mn} of

the fundamental response tensor. Consider, for example, the electric field response

$$\begin{aligned} (\chi_{EE}^p)^i_j(k, k') &= \frac{1}{\mu_0} \mathbb{D}_0(k) \left(k^0 \chi^i_j(k, k') k'_0 - k^i \chi^0_j(k, k') k'_0 \right. \\ &\quad \left. - k^0 \chi^i_0(k, k') k'_j + k^i \chi^0_0(k, k') k'_j \right) \mathbb{D}_0(k'). \end{aligned} \quad (5.48)$$

Using the constraints (5.9)–(5.11) on the fundamental response functions to express χ^0_j , χ^i_0 and χ^0_0 in terms of the spatial components χ_{mn} , we obtain

$$\begin{aligned} (\chi_{EE}^p)^i_j(\mathbf{k}, \mathbf{k}'; \omega) &= \frac{1}{\mu_0} \mathbb{D}_0(\mathbf{k}, \omega) \left(-\frac{\omega}{c} \delta_{im} + \frac{ck_i k_m}{\omega} \right) \chi_{mn}(\mathbf{k}, \mathbf{k}'; \omega) \\ &\quad \times \left(\frac{\omega}{c} \delta_{nj} - \frac{ck'_n k'_j}{\omega} \right) \mathbb{D}_0(\mathbf{k}', \omega). \end{aligned} \quad (5.49)$$

In the same way, we can express also the other partial derivatives in terms of χ_{mn} . We summarize all results by the following set of equations:

$$\frac{\delta E_{\text{ind}}^i(\mathbf{k}, \omega)}{\delta E_{\text{ext}}^j(\mathbf{k}', \omega)} = -\frac{1}{\varepsilon_0 \omega^2} \frac{\omega^2 \delta_{im} - c^2 k_i k_m}{\omega^2 - c^2 |\mathbf{k}|^2} \chi_{mn}(\mathbf{k}, \mathbf{k}'; \omega) \frac{\omega^2 \delta_{nj} - c^2 k'_n k'_j}{\omega^2 - c^2 |\mathbf{k}'|^2}, \quad (5.50)$$

$$\frac{1}{c} \frac{\delta E_{\text{ind}}^i(\mathbf{k}, \omega)}{\delta B_{\text{ext}}^j(\mathbf{k}', \omega)} = -\frac{1}{\varepsilon_0 \omega^2} \frac{\omega^2 \delta_{im} - c^2 k_i k_m}{\omega^2 - c^2 |\mathbf{k}|^2} \chi_{mn}(\mathbf{k}, \mathbf{k}'; \omega) \frac{\epsilon_{nlj} \omega ck'_\ell}{\omega^2 - c^2 |\mathbf{k}'|^2}, \quad (5.51)$$

$$c \frac{\delta B_{\text{ind}}^i(\mathbf{k}, \omega)}{\delta E_{\text{ext}}^j(\mathbf{k}', \omega)} = -\frac{1}{\varepsilon_0 \omega^2} \frac{\epsilon_{ikm} \omega ck_k}{\omega^2 - c^2 |\mathbf{k}|^2} \chi_{mn}(\mathbf{k}, \mathbf{k}'; \omega) \frac{\omega^2 \delta_{nj} - c^2 k'_n k'_j}{\omega^2 - c^2 |\mathbf{k}'|^2}, \quad (5.52)$$

$$\frac{\delta B_{\text{ind}}^i(\mathbf{k}, \omega)}{\delta B_{\text{ext}}^j(\mathbf{k}', \omega)} = -\frac{1}{\varepsilon_0 \omega^2} \frac{\epsilon_{ikm} \omega ck_k}{\omega^2 - c^2 |\mathbf{k}|^2} \chi_{mn}(\mathbf{k}, \mathbf{k}'; \omega) \frac{\epsilon_{nlj} \omega ck'_\ell}{\omega^2 - c^2 |\mathbf{k}'|^2}. \quad (5.53)$$

With the help of the electric and magnetic solution generators defined in

Section 4.1, these relations can be written compactly as

$$\overset{\leftrightarrow}{\chi}_{EE}^{\text{p}}(\mathbf{k}, \mathbf{k}'; \omega) = -\frac{1}{\varepsilon_0 \omega^2} \overset{\leftrightarrow}{E}(\mathbf{k}, \omega) \overset{\leftrightarrow}{\chi}(\mathbf{k}, \mathbf{k}'; \omega) \overset{\leftrightarrow}{E}(\mathbf{k}', \omega), \quad (5.54)$$

$$\overset{\leftrightarrow}{\chi}_{EB}^{\text{p}}(\mathbf{k}, \mathbf{k}'; \omega) = -\frac{1}{\varepsilon_0 \omega^2} \overset{\leftrightarrow}{E}(\mathbf{k}, \omega) \overset{\leftrightarrow}{\chi}(\mathbf{k}, \mathbf{k}'; \omega) \overset{\leftrightarrow}{B}(\mathbf{k}', \omega), \quad (5.55)$$

$$\overset{\leftrightarrow}{\chi}_{BE}^{\text{p}}(\mathbf{k}, \mathbf{k}'; \omega) = -\frac{1}{\varepsilon_0 \omega^2} \overset{\leftrightarrow}{B}(\mathbf{k}, \omega) \overset{\leftrightarrow}{\chi}(\mathbf{k}, \mathbf{k}'; \omega) \overset{\leftrightarrow}{E}(\mathbf{k}', \omega), \quad (5.56)$$

$$\overset{\leftrightarrow}{\chi}_{BB}^{\text{p}}(\mathbf{k}, \mathbf{k}'; \omega) = -\frac{1}{\varepsilon_0 \omega^2} \overset{\leftrightarrow}{B}(\mathbf{k}, \omega) \overset{\leftrightarrow}{\chi}(\mathbf{k}, \mathbf{k}'; \omega) \overset{\leftrightarrow}{B}(\mathbf{k}', \omega). \quad (5.57)$$

Thus, we have expressed all components of the field strength response tensor, i.e., the 36 partial derivatives of the induced electric/magnetic fields with respect to the external electric/magnetic fields, in terms of only 9 independent current response functions.

Finally, we remark that a covariant approach to the electromagnetic response functions has already been suggested by Hehl et al. [35, 68, 72]. The linear constitutive relations used in this approach, see [72, Eq. (15)], can be interpreted as

$$F_{\text{ext}}^{\mu\nu} = \tilde{\chi}^{\mu\nu}{}_{\alpha\beta} F_{\text{tot}}^{\alpha\beta}, \quad (5.58)$$

with a fourth-rank *constitutive tensor* $\tilde{\chi}$. These constitutive relations generalize the equation $\mathbf{D} = \varepsilon_0 \varepsilon_r \mathbf{E}$ to the case of linear bi-anisotropic materials [94]. Within linear response theory, the 36 components of the constitutive tensor are to be identified with the functional derivatives

$$\tilde{\chi}^{\mu\nu}{}_{\alpha\beta}(x, x') = \frac{\delta F_{\text{ext}}^{\mu\nu}(x)}{\delta F_{\text{tot}}^{\alpha\beta}(x')}. \quad (5.59)$$

Writing the total fields as a sum of the external and the induced fields, we see that

$$\frac{\delta F_{\text{tot}}^{\mu\nu}(x)}{\delta F_{\text{ext}}^{\alpha\beta}(x')} = \frac{1}{2} (\delta^\mu{}_\alpha \delta^\nu{}_\beta - \delta^\mu{}_\beta \delta^\nu{}_\alpha) \delta^4(x - x') + \frac{\delta F_{\text{ind}}^{\mu\nu}(x)}{\delta F_{\text{ext}}^{\alpha\beta}(x')}. \quad (5.60)$$

Consequently, the following formal relation holds between the constitutive tensor and the field strength response tensor:

$$(\tilde{\chi}^{-1})^{\mu\nu}{}_{\alpha\beta}(x, x') = \frac{1}{2} (\delta^\mu{}_\alpha \delta^\nu{}_\beta - \delta^\mu{}_\beta \delta^\nu{}_\alpha) \delta^4(x - x') + \chi^{\mu\nu}{}_{\alpha\beta}(x, x'), \quad (5.61)$$

where $\delta^\mu{}_\nu \equiv \eta^\mu{}_\nu$. This equation combined with Eq. (5.40) allows to express all 36 components of the constitutive tensor analytically in terms of only 9 independent components of the fundamental response tensor.

6. Universal response relations

6.1. Physical response functions

Within the linear regime, the electromagnetic properties of any material are typically characterized by the electric susceptibility tensor χ_e , which gives the polarization in terms of the total electric field,

$$\mathbf{P} = \varepsilon_0 \overset{\leftrightarrow}{\chi}_e \mathbf{E}, \quad (6.1)$$

by the magnetic susceptibility χ_m specifying the magnetization in terms of the external magnetic field,

$$\mathbf{M} = \overset{\leftrightarrow}{\chi}_m \mathbf{H}, \quad (6.2)$$

and by the conductivity σ giving the response of the induced current to the external electric field,²

$$\mathbf{j}_{\text{ind}} = \overset{\leftrightarrow}{\sigma} \mathbf{E}_{\text{ext}}. \quad (6.5)$$

Other common response functions are the relative permittivity or dielectric tensor ε_r defined by

$$\mathbf{D} = \varepsilon_0 \overset{\leftrightarrow}{\varepsilon}_r \mathbf{E}, \quad (6.6)$$

and the relative magnetic permeability μ_r defined by

$$\mathbf{B} = \mu_0 \overset{\leftrightarrow}{\mu}_r \mathbf{H}. \quad (6.7)$$

²The conductivity σ must not be confused with the *proper conductivity* $\tilde{\sigma}$, which is defined by

$$\mathbf{j}_{\text{ind}} = \tilde{\sigma} \mathbf{E}_{\text{tot}}. \quad (6.3)$$

These two notions of conductivity are related through

$$\tilde{\sigma} = \overset{\leftrightarrow}{\sigma} \overset{\leftrightarrow}{\varepsilon}_r, \quad (6.4)$$

with the dielectric tensor ε_r given by Eq. (6.6) (cf. [56]).

They are related to the corresponding susceptibilities by

$$\overset{\leftrightarrow}{\varepsilon}_{\text{r}} = 1 + \overset{\leftrightarrow}{\chi}_{\text{e}} , \quad (6.8)$$

$$\overset{\leftrightarrow}{\mu}_{\text{r}} = 1 + \overset{\leftrightarrow}{\chi}_{\text{m}} . \quad (6.9)$$

In principle, all these response functions represent 3×3 tensorial integral kernels with respect to the space and time variables.

Naively, one could expect that the components of the physical response tensors χ_{e} and χ_{m} correspond to functional derivatives which can be read out directly from the field strength response tensor of Section 5.4. It turns out, however, that this is not the case: although the components of the induced (and external) field strength tensors $F_{\text{ind}}^{\mu\nu}$ (and $F_{\text{ext}}^{\mu\nu}$) correspond to the induced (and external) electric and magnetic fields, which are observable, the components of the field strength *response* tensor $\chi^{\mu\nu}_{\alpha\beta}$ do not correspond to physical response functions (cf. [106]). In fact, the components of the field strength response tensor are not directly observable, and it is only the entire expansion (5.41) of the induced fields in terms of the external fields which has a physical meaning. The reason for this is that the field strength response tensor contains *partial* functional derivatives of the induced fields with respect to the external fields. Mathematically, such partial derivatives are obtained by varying the external electric and magnetic field independently of each other. In reality, however, the external electromagnetic fields have to fulfill the Maxwell equations and hence cannot be varied independently: partial derivatives do not correspond to physical perturbations.

Consider, for example, a typical physical response relation like Ohm's law,

$$\mathbf{j}_{\text{ind}}(\mathbf{k}, \omega) = \int d^3\mathbf{k}' \overset{\leftrightarrow}{\sigma}(\mathbf{k}, \mathbf{k}'; \omega) \mathbf{E}_{\text{ext}}(\mathbf{k}', \omega) , \quad (6.10)$$

where \mathbf{E}_{ext} is the external electric field acting on the medium. In general, a frequency-dependent electric field is also accompanied by a magnetic field \mathbf{B}_{ext} , and the actual current \mathbf{j}_{ind} in the medium is induced under the combined action of external electric and magnetic fields. Mathematically, this means that the measured conductivity has to be identified with the *total* functional derivative with respect to the external electric field:

$$\overset{\leftrightarrow}{\sigma}(\mathbf{k}, \mathbf{k}'; \omega) = \frac{d\mathbf{j}_{\text{ind}}(\mathbf{k}, \omega)}{d\mathbf{E}_{\text{ext}}(\mathbf{k}', \omega)} \quad (6.11)$$

$$\equiv \frac{\delta \mathbf{j}_{\text{ind}}(\mathbf{k}, \omega)}{\delta \mathbf{E}_{\text{ext}}(\mathbf{k}', \omega)} + \frac{\delta \mathbf{j}_{\text{ind}}(\mathbf{k}, \omega)}{\delta \mathbf{B}_{\text{ext}}(\mathbf{k}', \omega)} \frac{\delta \mathbf{B}_{\text{ext}}(\mathbf{k}', \omega)}{\delta \mathbf{E}_{\text{ext}}(\mathbf{k}', \omega)}. \quad (6.12)$$

By contrast, determining only the first term on the right hand side of this equation would require to introduce a hypothetical “partial” current which is induced exclusively by the external electric field. We conclude that only the total functional derivative is a directly observable response function. Moreover, we will see in the next subsection that the total functional derivative is directly related to the fundamental response tensor, which can be computed in the Kubo formalism. Mutatis mutandis, the same argument applies to any frequency-dependent *physical* (i.e. directly observable) response function, which should therefore be identified with a total functional derivative. In the following subsections, we will derive concrete expressions for the physical response functions and their general interrelations.

6.2. Optical conductivity

We first express the microscopic (frequency and wave-vector dependent) conductivity tensor in terms of the fundamental response tensor. In accordance with the above considerations, we define the microscopic conductivity tensor in real space as

$$\sigma_{k\ell}(\mathbf{x}, \mathbf{x}'; t - t') \stackrel{\text{def}}{=} \frac{dj_{\text{ind}}^k(\mathbf{x}, t)}{dE_{\text{ext}}^\ell(\mathbf{x}', t')}. \quad (6.13)$$

By a functional chain rule, this is equivalent to

$$\sigma_{k\ell}(\mathbf{x}, \mathbf{x}'; t - t') = c \int d^3\mathbf{y} \int ds \frac{\delta j_{\text{ind}}^k(\mathbf{x}, t)}{\delta A_{\text{ext}}^\mu(\mathbf{y}, s)} \frac{dA_{\text{ext}}^\mu(\mathbf{y}, s)}{dE_{\text{ext}}^\ell(\mathbf{x}', t')}. \quad (6.14)$$

We note that the total derivative has no bearing on the first term, because the induced current is per definitionem a functional of the external four-potential, while only the latter is regarded as a functional of the electromagnetic fields $\{\mathbf{E}_{\text{ext}}, \mathbf{B}_{\text{ext}}\}$. The functional derivative of the four-potential is calculated in the temporal gauge $\varphi_{\text{ext}} = 0$. Using the result (4.41) from Section 4.2, we obtain

$$\sigma_{k\ell}(\mathbf{x}, \mathbf{x}'; t - t') = - \int_{-\infty}^{\infty} ds \chi_{k\ell}(\mathbf{x}, \mathbf{x}'; t - s) \theta(s - t'). \quad (6.15)$$

By Fourier transforming with respect to the time variables, this is equivalent to

$$\sigma_{k\ell}(\mathbf{x}, \mathbf{x}'; \omega) = \frac{1}{i\omega} \chi_{k\ell}(\mathbf{x}, \mathbf{x}'; \omega). \quad (6.16)$$

In matrix notation, we can write this identity as

$$\overset{\leftrightarrow}{\sigma} = \frac{1}{i\omega} \overset{\leftrightarrow}{\chi}, \quad (6.17)$$

where

$$\overset{\leftrightarrow}{\chi} = \frac{\delta \mathbf{j}_{\text{ind}}}{\delta \mathbf{A}_{\text{ext}}} \quad (6.18)$$

is the spatial part of the fundamental response tensor (see Eq. (5.12)). In fact, Eq. (6.17) is a standard relation (see, e.g., [10, Section 3.4.4] or [107, Eq. (4.13)]), which in conjunction with the Kubo formula for the fundamental response tensor forms the basis for microscopic calculations of the optical conductivity [108–110]. We stress, however, that in this standard relation it is imperative to interpret the conductivity as the total derivative of the induced current with respect to the external electric field. A similar calculation of the *partial conductivity*

$$(\sigma^{\text{p}})_{k\ell}(\mathbf{k}, \mathbf{k}'; \omega) \stackrel{\text{def}}{=} \frac{\delta j_{\text{ind}}^k(\mathbf{k}, \omega)}{\delta E_{\text{ext}}^\ell(\mathbf{k}', \omega)} \quad (6.19)$$

yields instead the relation

$$(\sigma^{\text{p}})_{k\ell}(\mathbf{k}, \mathbf{k}'; \omega) = \frac{1}{i\omega} \chi_{km}(\mathbf{k}, \mathbf{k}'; \omega) \frac{\omega^2 \delta_{m\ell} - c^2 k'_m k'_\ell}{\omega^2 - c^2 |\mathbf{k}'|^2}. \quad (6.20)$$

Consequently, the partial and the total conductivity are related by

$$\overset{\leftrightarrow}{\sigma}^{\text{p}}(\mathbf{k}, \mathbf{k}'; \omega) = \overset{\leftrightarrow}{\sigma}(\mathbf{k}, \mathbf{k}'; \omega) \overset{\leftrightarrow}{\mathbb{E}}(\mathbf{k}', \omega), \quad (6.21)$$

where \mathbb{E} is the electric solution generator defined in Section 4.1. Finally, we note that by the constraints (5.9)–(5.11) on the fundamental response functions, we can express also χ^k_0 , χ^0_ℓ and χ^0_0 in terms of χ^k_ℓ and consequently in terms of $\sigma_{k\ell}$. Since χ^μ_ν can be reconstructed entirely from $\sigma_{k\ell}$, it follows that the microscopic conductivity tensor contains the complete information about the linear electromagnetic response (cf. [5]).

6.3. Dielectric tensor

We now derive the most general relation between the dielectric tensor (defined by Eq. (6.6)) and the microscopic conductivity tensor. The former determines the (magneto)optical properties of materials [111] and contains the entire information about elementary excitations and collective modes in solids [112]. With $\mathbf{E} \equiv \mathbf{E}_{\text{tot}} = \mathbf{E}_{\text{ext}} + \mathbf{E}_{\text{ind}}$ and $\mathbf{D} \equiv \varepsilon_0 \mathbf{E}_{\text{ext}}$, we have

$$[(\vec{\varepsilon}_r)^{-1}]_{ij}(x, x') = \frac{dE_{\text{tot}}^i(x)}{dE_{\text{ext}}^j(x')} \quad (6.22)$$

$$= \delta_{ij} \delta^4(x - x') + (\chi_{EE})_{ij}(x, x'), \quad (6.23)$$

where the second term determines the response of the induced to the external electric field. This is given by the fundamental relation (with $\mathbf{P} \equiv -\varepsilon_0 \mathbf{E}_{\text{ind}}$)

$$(\chi_{EE})_{ij}(x, x') = -\frac{1}{\varepsilon_0} \frac{dP^i(x)}{dE_{\text{ext}}^j(x')} \quad (6.24)$$

$$= -\frac{1}{\varepsilon_0} \int d^4y \int d^4y' \frac{\delta P^i(x)}{\delta j_{\text{ind}}^\mu(y)} \chi^\mu{}_\nu(y, y') \frac{dA_{\text{ext}}^\nu(y')}{dE_{\text{ext}}^j(x')}. \quad (6.25)$$

The first term in the integrand is evaluated as described in Section 5.3 by solving the equation of motion (5.25) for the induced field in terms of the induced sources: With the retarded Green function \mathcal{D}_0 for the scalar wave equation, we can express this as

$$\begin{aligned} P^i(\mathbf{x}, t) &= c \int d^3\mathbf{y} \int ds \frac{1}{\mu_0} \mathcal{D}_0(\mathbf{x} - \mathbf{y}, t - s) \\ &\times \left(\frac{\partial}{\partial y^i} \rho_{\text{ind}}(\mathbf{y}, s) + \frac{1}{c^2} \frac{\partial}{\partial s} j_{\text{ind}}^i(\mathbf{y}, s) \right). \end{aligned} \quad (6.26)$$

Hence after partial integration, we obtain the functional derivatives

$$\frac{1}{c} \frac{\delta P^i(\mathbf{x}, t)}{\delta \rho_{\text{ind}}(\mathbf{y}, s)} = -\frac{1}{c\mu_0} \frac{\partial}{\partial y^i} \mathcal{D}_0(\mathbf{x} - \mathbf{y}, t - s), \quad (6.27)$$

$$\frac{\delta P^i(\mathbf{x}, t)}{\delta j_{\text{ind}}^m(\mathbf{y}, s)} = -\delta_{im} \frac{1}{c^2\mu_0} \frac{\partial}{\partial s} \mathcal{D}_0(\mathbf{x} - \mathbf{y}, t - s). \quad (6.28)$$

For the last term in the integrand of Eq. (6.25) we use again Eq. (4.41). By putting these expressions into Eq. (6.25), performing partial integrations and

reexpressing the fundamental response tensor in terms of the conductivity tensor, we arrive at the relation

$$\begin{aligned}
[(\overset{\leftrightarrow}{\varepsilon}_r)^{-1}]_{ij}(\mathbf{x}, \mathbf{x}'; t - t') &= \frac{1}{c} \delta_{ij} \delta(t - t') \delta^3(\mathbf{x} - \mathbf{x}') \\
&- c \int d^3\mathbf{y} \int ds \int ds' \mathbb{D}_0(\mathbf{x} - \mathbf{y}; t - s) \\
&\times \left(\delta_{im} \frac{\partial^2}{\partial s^2} - c^2 \frac{\partial}{\partial y^i} \frac{\partial}{\partial y^m} \right) \sigma_{mj}(\mathbf{y}, \mathbf{x}'; s - s') \theta(s' - t').
\end{aligned} \tag{6.29}$$

This is the most general relation between the dielectric tensor and the microscopic conductivity tensor (assuming only homogeneity in time). By a Fourier transformation with respect to the space and time variables, it is equivalent to

$$\begin{aligned}
[(\overset{\leftrightarrow}{\varepsilon}_r)^{-1}]_{ij}(\mathbf{k}, \mathbf{k}'; \omega) &= \delta_{ij} \delta^3(\mathbf{k} - \mathbf{k}') \\
&- \frac{i}{\omega} \mathbb{D}_0(\mathbf{k}, \omega) (c^2 k_i k_m - \omega^2 \delta_{im}) \sigma_{mj}(\mathbf{k}, \mathbf{k}'; \omega).
\end{aligned} \tag{6.30}$$

In matrix notation, we can write this compactly as

$$\begin{aligned}
(\overset{\leftrightarrow}{\varepsilon}_r)^{-1}(\mathbf{k}, \mathbf{k}'; \omega) &= \overset{\leftrightarrow}{1} \delta^3(\mathbf{k} - \mathbf{k}') \\
&- \frac{i}{\omega} \mathbb{D}_0(\mathbf{k}, \omega) \left(c^2 |\mathbf{k}|^2 \overset{\leftrightarrow}{P}_L(\mathbf{k}) - \omega^2 \overset{\leftrightarrow}{1} \right) \overset{\leftrightarrow}{\sigma}(\mathbf{k}, \mathbf{k}'; \omega).
\end{aligned} \tag{6.31}$$

In Section 7.3 we will show that this general formula reduces to a well-known identity in the special case of homogeneous, isotropic materials and in a non-relativistic approximation.

6.4. Magnetic susceptibility

Next we consider the magnetic susceptibility, which contains the description of magnetic orderings and excitation spectra and is directly related to the neutron scattering cross section [113].³ In analogy to the electric case, we

³If χ^μ_ν is associated with the response of a microscopic *charge* current to the electromagnetic potential, the corresponding response function χ_m refers only to the *orbital* contribution to the magnetic susceptibility [114]. Spin contributions can be included in our picture by a divergence free contribution to the current of the Pauli equation, as it is motivated by the non-relativistic limit of the Dirac equation (cf. [115, chap. XX, §29]).

identify the physical response function with the total functional derivative

$$(\chi_m)_{ij}(x, x') = \mu_0 \frac{dM^i(x)}{dB_{\text{ext}}^j(x')} \quad (6.32)$$

$$= \mu_0 \int d^4y \int d^4y' \frac{\delta M^i(x)}{\delta j_{\text{ind}}^\mu(y)} \chi^\mu{}_\nu(y, y') \frac{dA_{\text{ext}}^\nu(y')}{dB_{\text{ext}}^j(x')}. \quad (6.33)$$

Here again, the total derivative displays its effect only in the functional derivative of the four-potential. Similarly as in the electric case, we obtain for the first term in the integrand

$$\frac{\delta M^i(\mathbf{x}, t)}{\delta j_{\text{ind}}^m(\mathbf{y}, s)} = -\frac{1}{\mu_0} \epsilon_{ikm} \frac{\partial}{\partial y^k} \mathbb{D}_0(\mathbf{x} - \mathbf{y}, t - s). \quad (6.34)$$

For the last term in the integrand of Eq. (6.33) we use the result (4.42) from Section 4.2. By putting these results into Eq. (6.33) and performing partial integrations with respect to the y and y' variables, we arrive at the relation

$$\begin{aligned} (\chi_m)_{ij}(\mathbf{x}, \mathbf{x}'; t - t') &= c \int d^3\mathbf{y} \int ds \int d^3\mathbf{y}' \int ds' \mathbb{D}_0(\mathbf{x} - \mathbf{y}; t - s) \\ &\times \left(\epsilon_{ikm} \epsilon_{j\ell n} \frac{\partial}{\partial y^k} \frac{\partial}{\partial y'^\ell} \chi_{mn}(\mathbf{y}, \mathbf{y}'; s - s') \right) \frac{1}{4\pi} \frac{\delta(s' - t')}{|\mathbf{y}' - \mathbf{x}'|}. \end{aligned} \quad (6.35)$$

By a Fourier transformation, this is equivalent to

$$(\chi_m)_{ij}(\mathbf{k}, \mathbf{k}'; \omega) = \mathbb{D}_0(\mathbf{k}, \omega) (\epsilon_{ikm} \epsilon_{j\ell n} k_k k'_\ell \chi_{mn}(\mathbf{k}, \mathbf{k}'; \omega)) \frac{1}{|\mathbf{k}'|^2}. \quad (6.36)$$

We will show in Section 7.3 that this general expression reduces to a well-known formula in the special case of homogeneous materials and in a non-relativistic approximation.

6.5. Magnetoelectric coupling and synopsis

For general materials, there are besides electric and magnetic susceptibilities also cross-coupling (magnetoelectric) coefficients, which specify the response of an induced magnetic field to an external electric field, and vice versa [94]. These can be calculated in close analogy to the examples above,

and we summarize all results by the following set of equations:

$$\frac{dE_{\text{ind}}^i(\mathbf{k}, \omega)}{dE_{\text{ext}}^j(\mathbf{k}', \omega)} = -\frac{1}{\varepsilon_0 \omega^2} \frac{\omega^2 \delta_{im} - c^2 k_i k_m}{\omega^2 - c^2 |\mathbf{k}|^2} i\omega \sigma_{mj}(\mathbf{k}, \mathbf{k}'; \omega), \quad (6.37)$$

$$\frac{1}{c} \frac{dE_{\text{ind}}^i(\mathbf{k}, \omega)}{dB_{\text{ext}}^j(\mathbf{k}', \omega)} = -\frac{1}{\varepsilon_0 \omega^2} \frac{\omega^2 \delta_{im} - c^2 k_i k_m}{\omega^2 - c^2 |\mathbf{k}|^2} i\omega \sigma_{mn}(\mathbf{k}, \mathbf{k}'; \omega) \frac{\epsilon_{nlj} \omega c k'_l}{-c^2 |\mathbf{k}'|^2}, \quad (6.38)$$

$$c \frac{dB_{\text{ind}}^i(\mathbf{k}, \omega)}{dE_{\text{ext}}^j(\mathbf{k}', \omega)} = -\frac{1}{\varepsilon_0 \omega^2} \frac{\epsilon_{ikm} \omega c k_k}{\omega^2 - c^2 |\mathbf{k}|^2} i\omega \sigma_{mj}(\mathbf{k}, \mathbf{k}'; \omega), \quad (6.39)$$

$$\frac{dB_{\text{ind}}^i(\mathbf{k}, \omega)}{dB_{\text{ext}}^j(\mathbf{k}', \omega)} = -\frac{1}{\varepsilon_0 \omega^2} \frac{\epsilon_{ikm} \omega c k_k}{\omega^2 - c^2 |\mathbf{k}|^2} i\omega \sigma_{mn}(\mathbf{k}, \mathbf{k}'; \omega) \frac{\epsilon_{nlj} \omega c k'_l}{-c^2 |\mathbf{k}'|^2}. \quad (6.40)$$

Using the electric and magnetic solution generators from Section 4.1, we can write these relations in matrix form as

$$\overset{\leftrightarrow}{\chi}_{EE}(\mathbf{k}, \mathbf{k}'; \omega) = -\frac{1}{\varepsilon_0 \omega^2} \overset{\leftrightarrow}{E}(\mathbf{k}, \omega) i\omega \overset{\leftrightarrow}{\sigma}(\mathbf{k}, \mathbf{k}'; \omega), \quad (6.41)$$

$$\overset{\leftrightarrow}{\chi}_{EB}(\mathbf{k}, \mathbf{k}'; \omega) = \overset{\leftrightarrow}{\chi}_{EE}(\mathbf{k}, \mathbf{k}'; \omega) \left(-\frac{\omega}{c |\mathbf{k}'|} \overset{\leftrightarrow}{R}_T(\mathbf{k}') \right), \quad (6.42)$$

$$\overset{\leftrightarrow}{\chi}_{BE}(\mathbf{k}, \mathbf{k}'; \omega) = -\frac{1}{\varepsilon_0 \omega^2} \overset{\leftrightarrow}{B}(\mathbf{k}, \omega) i\omega \overset{\leftrightarrow}{\sigma}(\mathbf{k}, \mathbf{k}'; \omega), \quad (6.43)$$

$$\overset{\leftrightarrow}{\chi}_{BB}(\mathbf{k}, \mathbf{k}'; \omega) = \overset{\leftrightarrow}{\chi}_{BE}(\mathbf{k}, \mathbf{k}'; \omega) \left(-\frac{\omega}{c |\mathbf{k}'|} \overset{\leftrightarrow}{R}_T(\mathbf{k}') \right). \quad (6.44)$$

To connect these physical response functions with those defined in Section 6.1, we note that the magnetic susceptibility coincides with (6.44), i.e.,

$$\overset{\leftrightarrow}{\chi}_m = \overset{\leftrightarrow}{\chi}_{BB}, \quad (6.45)$$

whereas the electric susceptibility is given in terms of (6.41) by

$$\overset{\leftrightarrow}{\chi}_e = -\overset{\leftrightarrow}{\chi}_{EE} (1 + \overset{\leftrightarrow}{\chi}_{EE})^{-1}. \quad (6.46)$$

For the permittivity and permeability tensors we further have

$$(\overset{\leftrightarrow}{\varepsilon}_{\text{r}})^{-1} = \overset{\leftrightarrow}{1} + \overset{\leftrightarrow}{\chi}_{EE}, \quad (6.47)$$

$$\overset{\leftrightarrow}{\mu}_{\text{r}} = \overset{\leftrightarrow}{1} + \overset{\leftrightarrow}{\chi}_{BB}. \quad (6.48)$$

The above relations between electromagnetic response functions are *universal*, i.e., valid on the microscopic and on the macroscopic scale and in any material, because of our model-independent definitions (1.9) and (1.12) of the electric and magnetic polarizations.⁴ Furthermore, the universal relations hold in any inertial frame, as they are derived from the Lorentz tensor of fundamental response functions. Given a particular microscopic model of the medium, Eqs. (6.41)–(6.44) can be evaluated in the Kubo formalism, where the conductivity tensor is expressed by a current-current correlation function [88, 96].

In concluding this subsection, we note that it is not yet obvious how the induced fields can be expressed in terms of the external perturbation by means of the physical response functions. The naive expansion, which would be analogous to the expansion (5.42)–(5.43) in terms of the partial derivatives, is in general not correct, i.e.,

$$\mathbf{E}_{\text{ind}} \neq \overset{\leftrightarrow}{\chi}_{EE} \mathbf{E}_{\text{ext}} + \overset{\leftrightarrow}{\chi}_{EB} c\mathbf{B}_{\text{ext}}, \quad (6.49)$$

$$c\mathbf{B}_{\text{ind}} \neq \overset{\leftrightarrow}{\chi}_{BE} \mathbf{E}_{\text{ext}} + \overset{\leftrightarrow}{\chi}_{BB} c\mathbf{B}_{\text{ext}}. \quad (6.50)$$

This is because the physical response functions correspond to total functional derivatives, hence part of the magnetic response is already contained in the electric response and vice versa. To solve this problem, we now take recourse to the canonical functional of Section 4.

6.6. Field expansions

In Section 4 we have introduced three different representations of the vector potential in the temporal gauge in terms of the electric and magnetic

⁴In applying the universal relations on a macroscopic scale, one must keep in mind the following limitation: Each macroscopic material property is usually dominated by a certain class of degrees of freedom. For example, the conductivity may be associated with the conduction electrons' orbital motion, whereas the magnetic susceptibility is often dominated by localized spin degrees of freedom. Thus, by describing each macroscopic response function only in terms of its most relevant degrees of freedom, the universal relations get lost. By contrast, the microscopic current response in principle refers to all degrees of freedom contributing to the electromagnetic four-current.

fields: (i) in terms of \mathbf{E} and \mathbf{B} as given by the canonical functional, (ii) only in terms of \mathbf{E} (and possibly a static magnetic field \mathbf{B}_0), and (iii) in terms of \mathbf{E}_L and \mathbf{B} (cf. Eqs. (4.60), (4.61) and (4.62)). As we are now going to show, these representations translate directly into three different electromagnetic field expansions. Consider, for example, the induced electric field, which can be expanded to first order in the external perturbation as

$$\mathbf{E}_{\text{ind}}(\mathbf{k}, \omega) = \frac{1}{\varepsilon_0} \frac{1}{i\omega} \overset{\leftrightarrow}{\mathbb{E}}(\mathbf{k}, \omega) \mathbf{j}_{\text{ind}}(\mathbf{k}, \omega) \quad (6.51)$$

$$= \frac{1}{\varepsilon_0} \frac{1}{i\omega} \overset{\leftrightarrow}{\mathbb{E}}(\mathbf{k}, \omega) \int d^3\mathbf{k}' \overset{\leftrightarrow}{\chi}(\mathbf{k}, \mathbf{k}'; \omega) \mathbf{A}_{\text{ext}}(\mathbf{k}', \omega). \quad (6.52)$$

Here \mathbf{A}_{ext} is the external vector potential in the temporal gauge, and we have used Eq. (4.23). Representing the external vector potential in terms of the electric and magnetic fields by the canonical functional (4.15), we obtain

$$\begin{aligned} \mathbf{E}_{\text{ind}}(\mathbf{k}, \omega) &= -\frac{1}{\varepsilon_0 \omega^2} \overset{\leftrightarrow}{\mathbb{E}}(\mathbf{k}, \omega) \int d^3\mathbf{k}' \overset{\leftrightarrow}{\chi}(\mathbf{k}, \mathbf{k}'; \omega) \\ &\quad \times \left\{ \overset{\leftrightarrow}{\mathbb{E}}(\mathbf{k}', \omega') \mathbf{E}_{\text{ext}}(\mathbf{k}', \omega) + \overset{\leftrightarrow}{\mathbb{B}}(\mathbf{k}', \omega') c\mathbf{B}_{\text{ext}}(\mathbf{k}', \omega) \right\} \end{aligned} \quad (6.53)$$

$$\begin{aligned} &= \int d^3\mathbf{k}' \overset{\leftrightarrow}{\chi}_{EE}^{\text{p}}(\mathbf{k}, \mathbf{k}'; \omega) \mathbf{E}_{\text{ext}}(\mathbf{k}', \omega) \\ &\quad + \int d^3\mathbf{k}' \overset{\leftrightarrow}{\chi}_{EB}^{\text{p}}(\mathbf{k}, \mathbf{k}'; \omega) c\mathbf{B}_{\text{ext}}(\mathbf{k}', \omega), \end{aligned} \quad (6.54)$$

where the partial functional derivatives are given by the expressions (5.54)–(5.55). On the other hand, using Eq. (4.30) to express \mathbf{A}_{ext} solely in terms of the electric field leads to

$$\mathbf{E}_{\text{ind}}(\mathbf{k}, \omega) = -\frac{1}{\varepsilon_0 \omega^2} \overset{\leftrightarrow}{\mathbb{E}}(\mathbf{k}, \omega) \int d^3\mathbf{k}' \overset{\leftrightarrow}{\chi}(\mathbf{k}, \mathbf{k}'; \omega) \mathbf{E}_{\text{ext}}(\mathbf{k}', \omega) \quad (6.55)$$

$$= \int d^3\mathbf{k}' \overset{\leftrightarrow}{\chi}_{EE}(\mathbf{k}, \mathbf{k}'; \omega) \mathbf{E}_{\text{ext}}(\mathbf{k}', \omega), \quad (6.56)$$

with the total functional derivative given by the universal relation (6.41). In the presence of a static magnetic field, the situation is slightly more complicated in that we earn the additional contribution

$$\int d^3\mathbf{k}' \overset{\leftrightarrow}{\chi}_{EB}(\mathbf{k}, \mathbf{k}'; \omega = 0) c\mathbf{B}_{\text{ext},0}(\mathbf{k}') \delta(\omega), \quad (6.57)$$

with $\mathbf{B}_{\text{ext},0}(\mathbf{x}) = \lim_{t \rightarrow -\infty} \mathbf{B}_{\text{ext}}(\mathbf{x}, t)$ (see Section 4.3). Finally, expressing \mathbf{A}_{ext} in Eq. (6.52) in terms of the longitudinal part of the electric field and the magnetic field as in Eq. (4.31) yields

$$\begin{aligned} \mathbf{E}_{\text{ind}}(\mathbf{k}, \omega) &= -\frac{1}{\varepsilon_0 \omega^2} \overset{\leftrightarrow}{E}(\mathbf{k}, \omega) \int d^3 \mathbf{k}' \overset{\leftrightarrow}{\chi}(\mathbf{k}, \mathbf{k}'; \omega) \\ &\times \left\{ (\mathbf{E}_{\text{ext}})_L(\mathbf{k}', \omega) + \left(-\frac{\omega}{c|\mathbf{k}'|} \right) \overset{\leftrightarrow}{R}_T(\mathbf{k}') c\mathbf{B}_{\text{ext}}(\mathbf{k}', \omega) \right\} \end{aligned} \quad (6.58)$$

$$\begin{aligned} &= \int d^3 \mathbf{k}' \overset{\leftrightarrow}{\chi}_{EE}(\mathbf{k}, \mathbf{k}'; \omega) (\mathbf{E}_{\text{ext}})_L(\mathbf{k}', \omega) \\ &+ \int d^3 \mathbf{k}' \overset{\leftrightarrow}{\chi}_{EB}(\mathbf{k}, \mathbf{k}'; \omega) c\mathbf{B}_{\text{ext}}(\mathbf{k}', \omega), \end{aligned} \quad (6.59)$$

where the cross-coupling tensor in the second line is given by the universal relation (6.42).

We conclude that as a matter of principle, there are three different but equivalent field expansions. First, in terms of the partial functional derivatives:

$$\mathbf{E}_{\text{ind}} = \overset{\leftrightarrow}{\chi}_{EE}^{\text{p}} \mathbf{E}_{\text{ext}} + \overset{\leftrightarrow}{\chi}_{EB}^{\text{p}} c\mathbf{B}_{\text{ext}}, \quad (6.60)$$

$$c\mathbf{B}_{\text{ind}} = \overset{\leftrightarrow}{\chi}_{BE}^{\text{p}} \mathbf{E}_{\text{ext}} + \overset{\leftrightarrow}{\chi}_{BB}^{\text{p}} c\mathbf{B}_{\text{ext}}. \quad (6.61)$$

This expansion is componentwise equivalent to the expansion (5.41) of the induced field strength tensor in terms of the external field strength tensor. Second, in terms of the external electric field, the static contribution to the magnetic field and the physical response functions:

$$\mathbf{E}_{\text{ind}} = \overset{\leftrightarrow}{\chi}_{EE} \mathbf{E}_{\text{ext}} + \overset{\leftrightarrow}{\chi}_{EB} c\mathbf{B}_{\text{ext},0}, \quad (6.62)$$

$$c\mathbf{B}_{\text{ind}} = \overset{\leftrightarrow}{\chi}_{BE} \mathbf{E}_{\text{ext}} + \overset{\leftrightarrow}{\chi}_{BB} c\mathbf{B}_{\text{ext},0}. \quad (6.63)$$

Third, we have a mixed expansion in terms of the longitudinal electric field, the magnetic field and the physical response functions:

$$\mathbf{E}_{\text{ind}} = \overset{\leftrightarrow}{\chi}_{EE} (\mathbf{E}_{\text{ext}})_L + \overset{\leftrightarrow}{\chi}_{EB} c\mathbf{B}_{\text{ext}}, \quad (6.64)$$

$$c\mathbf{B}_{\text{ind}} = \overset{\leftrightarrow}{\chi}_{BE} (\mathbf{E}_{\text{ext}})_L + \overset{\leftrightarrow}{\chi}_{BB} c\mathbf{B}_{\text{ext}}. \quad (6.65)$$

The total and the partial functional derivatives are related through functional chain rules, e.g.,

$$\overset{\leftrightarrow}{\chi}_{EE} = \frac{d\mathbf{E}_{\text{ind}}}{d\mathbf{E}_{\text{ext}}} = \frac{\delta\mathbf{E}_{\text{ind}}}{\delta\mathbf{E}_{\text{ext}}} + \frac{\delta\mathbf{E}_{\text{ind}}}{\delta\mathbf{B}_{\text{ext}}} \frac{\delta\mathbf{B}_{\text{ext}}}{\delta\mathbf{E}_{\text{ext}}} \quad (6.66)$$

$$= \overset{\leftrightarrow}{\chi}_{EE}^{\text{p}} + \overset{\leftrightarrow}{\chi}_{EB}^{\text{p}} \left(\frac{c|\mathbf{k}'|}{\omega} \overset{\leftrightarrow}{R}_{\text{T}}(\mathbf{k}') \right), \quad (6.67)$$

and similarly,

$$\overset{\leftrightarrow}{\chi}_{EB} = \overset{\leftrightarrow}{\chi}_{EE}^{\text{p}} \left(-\frac{\omega}{c|\mathbf{k}'|} \overset{\leftrightarrow}{R}_{\text{T}}(\mathbf{k}') \right) + \overset{\leftrightarrow}{\chi}_{EB}^{\text{p}}. \quad (6.68)$$

Here we have used Eqs. (4.37) and (4.38) respectively. We remark that for *longitudinal* electric perturbations, the second term in Eq. (6.67) vanishes and therefore,

$$\mathbf{E}_{\text{ind}} = \overset{\leftrightarrow}{\chi}_{EE}(\mathbf{E}_{\text{ext}})_{\text{L}} = \overset{\leftrightarrow}{\chi}_{EE}^{\text{p}}(\mathbf{E}_{\text{ext}})_{\text{L}}. \quad (6.69)$$

Hence in this case the expansions in terms of the total and the partial functional derivatives coincide.

6.7. Magnetic conductivity

To complete our discussion of the physical response functions, we now turn our attention to the induced current \mathbf{j}_{ind} , which can be expanded in an analogous way in terms of the external electric and magnetic fields. We define the *magnetic conductivity* as

$$\overset{\leftrightarrow}{\kappa} = \frac{1}{c} \frac{d\mathbf{j}_{\text{ind}}}{d\mathbf{B}_{\text{ext}}}. \quad (6.70)$$

This is related to the fundamental response functions by

$$\overset{\leftrightarrow}{\kappa}(\mathbf{k}, \mathbf{k}'; \omega) = \frac{1}{c} \frac{\delta\mathbf{j}_{\text{ind}}(\mathbf{k}, \omega)}{\delta\mathbf{A}_{\text{ext}}(\mathbf{k}', \omega)} \frac{d\mathbf{A}_{\text{ext}}(\mathbf{k}', \omega)}{d\mathbf{B}_{\text{ext}}(\mathbf{k}', \omega)} \quad (6.71)$$

$$= \overset{\leftrightarrow}{\chi}(\mathbf{k}, \mathbf{k}'; \omega) \frac{1}{i\omega} \left(-\frac{\omega}{c|\mathbf{k}'|} \overset{\leftrightarrow}{R}_{\text{T}}(\mathbf{k}') \right). \quad (6.72)$$

Consequently, the following universal relation holds:

$$\overset{\leftrightarrow}{\kappa}(\mathbf{k}, \mathbf{k}'; \omega) = \overset{\leftrightarrow}{\sigma}(\mathbf{k}, \mathbf{k}'; \omega) \left(-\frac{\omega}{c|\mathbf{k}'|} \overset{\leftrightarrow}{R}_{\text{T}}(\mathbf{k}') \right). \quad (6.73)$$

The corresponding *partial* magnetic conductivity is defined by

$$\overset{\leftrightarrow}{\kappa}^{\text{p}} = \frac{1}{c} \frac{\delta \mathbf{j}_{\text{ind}}}{\delta \mathbf{B}_{\text{ext}}} . \quad (6.74)$$

The partial electric and magnetic conductivities can be expressed in terms of the electric and magnetic solution generators from Section 4.1 as

$$\overset{\leftrightarrow}{\sigma}^{\text{p}}(\mathbf{k}, \mathbf{k}'; \omega) = \frac{1}{i\omega} \overset{\leftrightarrow}{\chi}(\mathbf{k}, \mathbf{k}'; \omega) \overset{\leftrightarrow}{E}(\mathbf{k}', \omega) , \quad (6.75)$$

$$\overset{\leftrightarrow}{\kappa}^{\text{p}}(\mathbf{k}, \mathbf{k}'; \omega) = \frac{1}{i\omega} \overset{\leftrightarrow}{\chi}(\mathbf{k}, \mathbf{k}'; \omega) \overset{\leftrightarrow}{B}(\mathbf{k}', \omega) . \quad (6.76)$$

Analogous to the case of the induced electromagnetic fields, the following three different but equivalent expansions of the induced current hold to first order in the external perturbation:

$$\mathbf{j}_{\text{ind}} = \overset{\leftrightarrow}{\sigma}^{\text{p}} \mathbf{E}_{\text{ext}} + \overset{\leftrightarrow}{\kappa}^{\text{p}} c \mathbf{B}_{\text{ext}} \quad (6.77)$$

$$= \overset{\leftrightarrow}{\sigma} \mathbf{E}_{\text{ext}} + \overset{\leftrightarrow}{\kappa} c \mathbf{B}_{\text{ext},0} \quad (6.78)$$

$$= \overset{\leftrightarrow}{\sigma} (\mathbf{E}_{\text{ext}})_{\text{L}} + \overset{\leftrightarrow}{\kappa} c \mathbf{B}_{\text{ext}} . \quad (6.79)$$

In this case, however, the second expansion (6.78) is practically preferred: It induces a decomposition of the induced current into a dynamical part responding to the external electric field and a temporally constant current $\mathbf{j}_{\text{ind},0}$ induced by a static magnetic field,

$$\mathbf{j}_{\text{ind},0} = \overset{\leftrightarrow}{\kappa} c \mathbf{B}_{\text{ext},0} . \quad (6.80)$$

It is natural to split off this contribution from the dynamical current by making the transition to the corresponding static magnetization \mathbf{M}_0 , which is defined by

$$\nabla \times \mathbf{M}_0 = \mathbf{j}_{\text{ind},0} , \quad (6.81)$$

$$\nabla \cdot \mathbf{M}_0 = 0 . \quad (6.82)$$

Microscopically, such a transition can be motivated by the fact that often the magnetization is dominated by the spin degrees of freedom, while the dynamical current is dominated by the conduction electron's orbital motion.

The response of \mathbf{M}_0 to the static external magnetic field is now given by the instantaneous limit of the magnetic susceptibility,

$$\mathbf{M}_0 = \frac{\overleftrightarrow{\chi}_m(\omega=0)}{\mu_0} \mathbf{B}_{\text{ext},0}. \quad (6.83)$$

Here, χ_m is related to the magnetic conductivity by

$$\overleftrightarrow{\chi}_m(\mathbf{k}, \mathbf{k}'; \omega) = \frac{1}{\varepsilon_0} \frac{1}{i\omega} \overleftrightarrow{B}(\mathbf{k}, \omega) \overleftrightarrow{\kappa}(\mathbf{k}, \mathbf{k}'; \omega) \quad (6.84)$$

$$= i\mathcal{D}_0(\mathbf{k}, \omega) c|\mathbf{k}| \overleftrightarrow{R}_T(\mathbf{k}) \overleftrightarrow{\kappa}(\mathbf{k}, \mathbf{k}'; \omega), \quad (6.85)$$

which follows by comparing the universal relations for χ_m and κ , Eq. (6.45) together with (6.43)–(6.44) and Eq. (6.73), respectively. The remaining dynamical current then fulfills exactly

$$\dot{\mathbf{j}}_{\text{ind}} - \dot{\mathbf{j}}_{\text{ind},0} = \overleftrightarrow{\sigma} \mathbf{E}_{\text{ext}}. \quad (6.86)$$

This shows that to first order in the external perturbation, Ohm's law can be upheld in the presence of magnetic fields. In fact, Ohm's law is even relativistically covariant (see [95] for a discussion).

7. Empirical limiting cases

In their most general form, the universal response relations (6.41)–(6.44) are not always particularly useful. In practice, one should take them to several empirically motivated limits. Foremost among these are:

- (i) The *homogeneous limit*, where response functions are proportional to $\delta^3(\mathbf{k} - \mathbf{k}')$ and hence essentially depend only on one momentum,
- (ii) the *isotropic limit*, where spatial response tensors are of the form

$$\overleftrightarrow{\chi}(\mathbf{k}, \omega) = \chi_L(\mathbf{k}, \omega) \overleftrightarrow{P}_L(\mathbf{k}) + \chi_T(\mathbf{k}, \omega) \overleftrightarrow{P}_T(\mathbf{k}) \quad (7.1)$$

with longitudinal and transverse response functions χ_L and χ_T ,

- (iii) the *ultra-relativistic limit*, where $\omega = c|\mathbf{k}|$,
- (iv) the *non-relativistic limit*, where $\omega \ll c|\mathbf{k}|$,

(v) the *instantaneous limit*, where $\omega = 0$.

In this section, we will derive concrete expressions for both the partial functional derivatives of Section 5.4 and the physical response functions of Section 6 in these limiting cases.

7.1. Homogeneous and isotropic limit

We equate $\mathbf{k} = \mathbf{k}'$ in the expressions (5.54)–(5.57) for the partial functional derivatives and assume an isotropic current response as described by Eq. (7.1). By using the algebra of the projection and rotation operators shown in Table 1, the components of the field strength response tensor then simplify as

$$\overset{\leftrightarrow}{\chi}_{EE}^{\text{p}}(\mathbf{k}, \omega) = -\frac{1}{\varepsilon_0 \omega^2} \chi_{\text{L}}(\mathbf{k}, \omega) \overset{\leftrightarrow}{P}_{\text{L}}(\mathbf{k}) - \overset{\leftrightarrow}{\chi}_{BB}^{\text{p}}(\mathbf{k}, \omega), \quad (7.2)$$

$$\overset{\leftrightarrow}{\chi}_{EB}^{\text{p}}(\mathbf{k}, \omega) = \overset{\leftrightarrow}{\chi}_{BE}^{\text{p}}(\mathbf{k}, \omega), \quad (7.3)$$

$$\overset{\leftrightarrow}{\chi}_{BE}^{\text{p}}(\mathbf{k}, \omega) = -\varepsilon_0 \omega^2 \mathbb{D}_0(\mathbf{k}, \omega) \frac{c|\mathbf{k}|}{\omega} \chi_{\text{T}}(\mathbf{k}, \omega) \mathbb{D}_0(\mathbf{k}, \omega) \overset{\leftrightarrow}{R}_{\text{T}}(\mathbf{k}), \quad (7.4)$$

$$\overset{\leftrightarrow}{\chi}_{BB}^{\text{p}}(\mathbf{k}, \omega) = \varepsilon_0 \omega^2 \mathbb{D}_0(\mathbf{k}, \omega) \frac{c^2|\mathbf{k}|^2}{\omega^2} \chi_{\text{T}}(\mathbf{k}, \omega) \mathbb{D}_0(\mathbf{k}, \omega) \overset{\leftrightarrow}{P}_{\text{T}}(\mathbf{k}). \quad (7.5)$$

Similarly, the universal response relations (6.41)–(6.44) reduce to

$$\overset{\leftrightarrow}{\chi}_{EE}(\mathbf{k}, \omega) = -\frac{1}{\varepsilon_0 \omega^2} \chi_{\text{L}}(\mathbf{k}, \omega) \overset{\leftrightarrow}{P}_{\text{L}}(\mathbf{k}) + \overset{\leftrightarrow}{\chi}_{BB}(\mathbf{k}, \omega), \quad (7.6)$$

$$\overset{\leftrightarrow}{\chi}_{EB}(\mathbf{k}, \omega) = -\mathbb{D}_0(\mathbf{k}, \omega) \frac{\omega}{c|\mathbf{k}|} \chi_{\text{T}}(\mathbf{k}, \omega) \overset{\leftrightarrow}{R}_{\text{T}}(\mathbf{k}), \quad (7.7)$$

$$\overset{\leftrightarrow}{\chi}_{BE}(\mathbf{k}, \omega) = \mathbb{D}_0(\mathbf{k}, \omega) \frac{c|\mathbf{k}|}{\omega} \chi_{\text{T}}(\mathbf{k}, \omega) \overset{\leftrightarrow}{R}_{\text{T}}(\mathbf{k}), \quad (7.8)$$

$$\overset{\leftrightarrow}{\chi}_{BB}(\mathbf{k}, \omega) = \mathbb{D}_0(\mathbf{k}, \omega) \chi_{\text{T}}(\mathbf{k}, \omega) \overset{\leftrightarrow}{P}_{\text{T}}(\mathbf{k}). \quad (7.9)$$

For later purposes, we further note that in the homogeneous and isotropic

limit the *density response function*

$$\chi = \frac{\delta \rho_{\text{ind}}}{\delta \varphi_{\text{ext}}} = \frac{1}{c^2} \chi^0_0 \quad (7.10)$$

is related to the longitudinal current response function χ_L by

$$\chi(\mathbf{k}, \omega) = -\frac{k_i k_j}{\omega^2} \chi_{ij}(\mathbf{k}, \omega) = -\frac{|\mathbf{k}|^2}{\omega^2} \chi_L(\mathbf{k}, \omega), \quad (7.11)$$

as follows from the representation (5.12) of the fundamental response tensor. In the following subsections, we will discuss the ultra-relativistic limit $\omega = c|\mathbf{k}|$, the non-relativistic limit $\omega \ll c|\mathbf{k}|$ and the instantaneous limit $\omega = 0$ presuming homogeneity and isotropy.

7.2. Ultra-relativistic limit

In the ultra-relativistic limit where $\omega = c|\mathbf{k}|$, the universal response relations read as follows:

$$\overleftrightarrow{\chi}_{EE}(\mathbf{k}, \omega) = \overleftrightarrow{\chi}_{BB}(\mathbf{k}, \omega), \quad (7.12)$$

$$\overleftrightarrow{\chi}_{EB}(\mathbf{k}, \omega) = -\overleftrightarrow{\chi}_{BE}(\mathbf{k}, \omega), \quad (7.13)$$

$$\overleftrightarrow{\chi}_{BE}(\mathbf{k}, \omega) = \mathbb{D}_0(\mathbf{k}, \omega) \chi_T(\mathbf{k}, \omega) \overleftrightarrow{R}_T(\mathbf{k}), \quad (7.14)$$

$$\overleftrightarrow{\chi}_{BB}(\mathbf{k}, \omega) = \mathbb{D}_0(\mathbf{k}, \omega) \chi_T(\mathbf{k}, \omega) \overleftrightarrow{P}_T(\mathbf{k}). \quad (7.15)$$

These equations can formally be obtained by the replacement $\omega \mapsto c|\mathbf{k}|$ in Eqs. (7.6)–(7.9) and by omitting the first term on the right hand side of Eq. (7.6). However, as the response functions are singular distributions and hence only their action on (external) fields is well-defined, we must discuss these formulae more thoroughly: The dispersion relation $\omega = c|\mathbf{k}|$ means that the external fields are vacuum solutions of the Maxwell equations, which are purely transverse. In particular, this implies that $\rho_{\text{ext}} = \varepsilon_0 \nabla \cdot \mathbf{E}_{\text{ext}} = 0$. By isotropy, the induced fields are also transverse, and consequently the longitudinal electric field response vanishes in the ultra-relativistic limit. The induced fields satisfy the equations of motion

$$\square \mathbf{E}_{\text{ind}} = -\mu_0 \partial_t \mathbf{j}_{\text{ind}}, \quad (7.16)$$

$$\square \mathbf{B}_{\text{ind}} = \mu_0 \nabla \times \mathbf{j}_{\text{ind}}, \quad (7.17)$$

where we have used $\rho_{\text{ind}} = 0$ by the transversality of the induced electric field. With $\mathbf{E}_{\text{ext}} = -\partial_t \mathbf{A}_{\text{ext}}$, the vector potential \mathbf{A}_{ext} is also transverse. Hence the induced current is given to linear order by

$$\mathbf{j}_{\text{ind}} = \chi_{\text{T}} \mathbf{A}_{\text{ext}} , \quad (7.18)$$

with the transverse response function χ_{T} . Putting this result into Eqs. (7.16)–(7.17), performing partial integrations and reexpressing \mathbf{A}_{ext} in terms of the electric and magnetic fields yields

$$\square \mathbf{E}_{\text{ind}} = \mu_0 \chi_{\text{T}} \mathbf{E}_{\text{ext}} , \quad (7.19)$$

$$\square \mathbf{B}_{\text{ind}} = \mu_0 \chi_{\text{T}} \mathbf{B}_{\text{ext}} . \quad (7.20)$$

Using the retarded Green function \mathcal{D}_0 of the scalar wave equation, these equations can be inverted as

$$\mathbf{E}_{\text{ind}} = \mathcal{D}_0 \chi_{\text{T}} \mathbf{E}_{\text{ext}} , \quad (7.21)$$

$$\mathbf{B}_{\text{ind}} = \mathcal{D}_0 \chi_{\text{T}} \mathbf{B}_{\text{ext}} . \quad (7.22)$$

Thus, we have proven Eqs. (7.12) and (7.15). Furthermore, for $\omega = c|\mathbf{k}|$ the external electric and magnetic fields are related by Faraday's law as

$$c\mathbf{B} = \frac{c\mathbf{k} \times \mathbf{E}}{\omega} = \frac{\mathbf{k} \times \mathbf{E}}{|\mathbf{k}|} = \overset{\leftrightarrow}{R}_{\text{T}}(\mathbf{k}) \mathbf{E} , \quad (7.23)$$

or equivalently,

$$\mathbf{E} = -\overset{\leftrightarrow}{R}_{\text{T}}(\mathbf{k}) c\mathbf{B} . \quad (7.24)$$

Therefore, we obtain also the relations

$$\mathbf{E}_{\text{ind}} = -\mathcal{D}_0 \chi_{\text{T}} \overset{\leftrightarrow}{R}_{\text{T}}(\mathbf{k}) c\mathbf{B}_{\text{ext}} , \quad (7.25)$$

$$c\mathbf{B}_{\text{ind}} = \mathcal{D}_0 \chi_{\text{T}} \overset{\leftrightarrow}{R}_{\text{T}}(\mathbf{k}) \mathbf{E}_{\text{ext}} , \quad (7.26)$$

which yield Eqs. (7.13)–(7.14) for the cross-coupling coefficients. We note that in the ultra-relativistic limit, the redundancy of the field expansion in

terms of the physical response functions becomes particularly evident. In fact, we have

$$\mathbf{E}_{\text{ind}} = \overset{\leftrightarrow}{\chi}_{EE} \mathbf{E}_{\text{ext}} = \overset{\leftrightarrow}{\chi}_{EB} c \mathbf{B}_{\text{ext}}, \quad (7.27)$$

$$c \mathbf{B}_{\text{ind}} = \overset{\leftrightarrow}{\chi}_{BE} \mathbf{E}_{\text{ext}} = \overset{\leftrightarrow}{\chi}_{BB} c \mathbf{B}_{\text{ext}}, \quad (7.28)$$

instead of the naive expansion (6.49)–(6.50) in terms of \mathbf{E}_{ext} and \mathbf{B}_{ext} .

We remark that the partial functional derivatives become ill-defined in the ultra-relativistic limit. The reason for this is that the partial derivatives rely on the canonical functional (4.16), which had been constructed for retarded fields generated by sources and therefore does not apply to vacuum fields. Correspondingly, Eq. (4.16) is formally divergent at $\omega = c|\mathbf{k}|$. By contrast, as argued in Section 4.2, the functionals (4.28) and (4.29), on which the physical response functions are based, are valid in the vacuum as well and can be evaluated at $\omega = c|\mathbf{k}|$.

7.3. Non-relativistic limit

The non-relativistic limit where $\omega \ll c|\mathbf{k}|$ is treated by approximating the scalar Green function $\mathcal{D}_0(\mathbf{k}, \omega)$ by its zero-frequency limit,

$$\mathcal{D}_0(\mathbf{k}, \omega = 0) = \frac{\mu_0}{|\mathbf{k}|^2} \equiv \frac{v(\mathbf{k})}{c^2}, \quad (7.29)$$

while keeping the frequency dependence of the fundamental response functions. Here, $v(\mathbf{k})$ denotes the Coulomb potential given by

$$v(\mathbf{k}) \equiv v(\mathbf{k}, \omega) = \frac{1}{\varepsilon_0 |\mathbf{k}|^2}, \quad (7.30)$$

or in real space by

$$v(\mathbf{x} - \mathbf{x}', t - t') = \frac{1}{4\pi\varepsilon_0} \frac{\delta(ct - ct')}{|\mathbf{x} - \mathbf{x}'|}. \quad (7.31)$$

Hence, in this limit relativistic retardation effects are neglected. The non-relativistic limit is especially interesting in the case of the dielectric tensor. In a homogeneous medium and for $\omega \ll c|\mathbf{k}|$, Eq. (6.31) simplifies as

$$(\overset{\leftrightarrow}{\varepsilon}_{\text{r}})^{-1}(\mathbf{k}, \omega) = \overset{\leftrightarrow}{1} - \frac{\text{i}}{\omega\varepsilon_0} \overset{\leftrightarrow}{P}_{\text{L}}(\mathbf{k}) \overset{\leftrightarrow}{\sigma}(\mathbf{k}, \omega). \quad (7.32)$$

Specializing to an isotropic medium, where

$$\overset{\leftrightarrow}{\sigma}(\mathbf{k}, \omega) = \sigma_L(\mathbf{k}, \omega) \overset{\leftrightarrow}{P}_L(\mathbf{k}) + \sigma_T(\mathbf{k}, \omega) \overset{\leftrightarrow}{P}_T(\mathbf{k}) \quad (7.33)$$

with longitudinal and transverse conductivities σ_L and σ_T , we obtain the simpler relation

$$(\overset{\leftrightarrow}{\varepsilon}_r)^{-1}(\mathbf{k}, \omega) = \overset{\leftrightarrow}{1} - \frac{i}{\omega \varepsilon_0} \sigma_L(\mathbf{k}, \omega) \overset{\leftrightarrow}{P}_L(\mathbf{k}). \quad (7.34)$$

In particular, the *longitudinal dielectric constant* $\varepsilon_{r,L}$ is related to the longitudinal conductivity by

$$\varepsilon_{r,L}^{-1}(\mathbf{k}, \omega) = 1 - \frac{i}{\omega \varepsilon_0} \sigma_L(\mathbf{k}, \omega). \quad (7.35)$$

This is indeed a well-known identity [9, Eq. (6.51)]. We have shown that it follows from the general relation (6.31) in a non-relativistic approximation and by restricting to homogeneous, isotropic materials.

Similarly, for the magnetic susceptibility χ_m we obtain from Eq. (6.36) in the homogeneous and non-relativistic limit

$$(\chi_m)_{ij}(\mathbf{k}, \omega) = \mu_0 \frac{1}{|\mathbf{k}|^4} \epsilon_{ikm} \epsilon_{j\ell n} k_k k_\ell \chi_{mn}(\mathbf{k}, \omega). \quad (7.36)$$

This formula agrees for $\omega = 0$ with [116, Eq. (AI.6)]. In isotropic media, it further simplifies as

$$(\chi_m)_{ij}(\mathbf{k}, \omega) = \mu_0 \frac{\chi_T(\mathbf{k}, \omega)}{|\mathbf{k}|^2} \left(\delta_{ij} - \frac{k_i k_j}{|\mathbf{k}|^2} \right). \quad (7.37)$$

Since χ_m acts on magnetic fields, which are purely transverse anyway, the transverse projection operator in brackets can be omitted and we arrive at

$$\chi_m(\mathbf{k}, \omega) = \mu_0 \frac{\chi_T(\mathbf{k}, \omega)}{|\mathbf{k}|^2}. \quad (7.38)$$

The two equations (7.35) and (7.38) can be written in an analogous way if we express the longitudinal conductivity σ_L through the longitudinal current response function χ_L . We then get the relations for the longitudinal

permittivity and the permeability

$$\varepsilon_{\text{r,L}}^{-1}(\mathbf{k}, \omega) = 1 - \frac{1}{\varepsilon_0 \omega^2} \chi_{\text{L}}(\mathbf{k}, \omega), \quad (7.39)$$

$$\mu_{\text{r}}(\mathbf{k}, \omega) = 1 + \frac{\mu_0}{|\mathbf{k}|^2} \chi_{\text{T}}(\mathbf{k}, \omega). \quad (7.40)$$

Further relating χ_{L} to the density response function χ via Eq. (7.11) and using Eq. (7.30), we arrive at the standard formulae [9, 10]

$$\varepsilon_{\text{r,L}}^{-1}(\mathbf{k}, \omega) = 1 + v(\mathbf{k}) \chi(\mathbf{k}, \omega), \quad (7.41)$$

$$\mu_{\text{r}}(\mathbf{k}, \omega) = 1 + \frac{1}{c^2} v(\mathbf{k}) \chi_{\text{T}}(\mathbf{k}, \omega). \quad (7.42)$$

Finally, we comment on the cross-coupling coefficients in the non-relativistic limit. One sees immediately from Eq. (7.7) that χ_{EB} becomes small in this limit due to the factor $\omega/c|\mathbf{k}|$. By contrast, χ_{BE} becomes large because of the inverse factor in Eq. (7.8). However, this does not have any physical consequences, because in the expansion

$$c\mathbf{B}_{\text{ind}} = \overset{\leftrightarrow}{\chi}_{BE}(\mathbf{E}_{\text{ext}})_{\text{L}} + \overset{\leftrightarrow}{\chi}_{BB} c\mathbf{B}_{\text{ext}}, \quad (7.43)$$

this response function acts on the longitudinal electric field. As χ_{BE} contains the transverse rotation operator, this yields zero.

7.4. Instantaneous limit

As mentioned in Section 2.1, the limit $\omega \rightarrow 0$ corresponds to static (induced and external) field quantities and to instantaneous response functions. Consider again Eqs. (7.6)–(7.9) for the physical response functions in the homogeneous and isotropic limit. By Faraday's law, static electric fields are purely longitudinal. Since χ_{BE} contains the transverse rotation operator, this response function effectively vanishes. For χ_{EB} the limit $\omega \rightarrow 0$ can be performed trivially giving also $\chi_{EB} = 0$. The instantaneous limit of the longitudinal dielectric constant $\varepsilon_{\text{r,L}}$ and the magnetic susceptibility χ_{m} can be inferred from Eqs. (7.41)–(7.42) in the previous subsection, where it remains to set $\omega = 0$. In particular, we recover the well-known formula for

the (orbital) magnetic susceptibility [10, Eq. (3.183)],

$$\chi_m = \mu_0 \lim_{|\mathbf{k}| \rightarrow 0} \frac{\chi_T(\mathbf{k}, \omega = 0)}{|\mathbf{k}|^2}, \quad (7.44)$$

where the limit $|\mathbf{k}| \rightarrow 0$ corresponds to integrating out the spatial dependence. We conclude that in the homogeneous, isotropic and instantaneous limit the universal response relations simplify as

$$\overset{\leftrightarrow}{\chi}_{EE}(\mathbf{k}, 0) = v(\mathbf{k}) \chi(\mathbf{k}, 0) \overset{\leftrightarrow}{P}_L(\mathbf{k}), \quad (7.45)$$

$$\overset{\leftrightarrow}{\chi}_{EB}(\mathbf{k}, 0) = 0, \quad (7.46)$$

$$\overset{\leftrightarrow}{\chi}_{BE}(\mathbf{k}, 0) = 0, \quad (7.47)$$

$$\overset{\leftrightarrow}{\chi}_{BB}(\mathbf{k}, 0) = \frac{1}{c^2} v(\mathbf{k}) \chi_T(\mathbf{k}, 0) \overset{\leftrightarrow}{P}_T(\mathbf{k}). \quad (7.48)$$

Since static electric fields are always longitudinal and magnetic fields are always transverse, we may omit the projection operators in Eqs. (7.45) and (7.48) and write shorthand

$$\chi_{EE}(\mathbf{k}) = v(\mathbf{k}) \chi(\mathbf{k}), \quad (7.49)$$

$$\chi_{BB}(\mathbf{k}) = \frac{1}{c^2} v(\mathbf{k}) \chi_T(\mathbf{k}). \quad (7.50)$$

Thus, in the instantaneous limit the medium is effectively described by only two independent scalar response functions. These relate the induced electric or magnetic field to the external electric or magnetic field respectively, while the cross couplings vanish. The reason for this is indeed simple: if the response function is isotropic, a static (i.e. longitudinal) electric field can only induce a longitudinal electric field but not a transverse magnetic field, and the converse holds true for a magnetic (i.e. transverse) perturbation. Finally, we note that the same expressions (7.45)–(7.48) can be derived even more directly for the partial response functions starting from Eqs. (7.2)–(7.5), which means that in the instantaneous limit the total and the partial derivatives coincide. This is again consistent with the fact that static electric fields are purely longitudinal and hence decouple completely from the transverse magnetic fields.

8. Conclusion

The Functional Approach developed in this paper disentangles electrodynamics of media from macroscopic electrodynamics: while the latter corresponds to spatial and/or temporal averaging of the fields (which can be performed with or without media), electrodynamics of media is set up by regarding induced quantities as functionals of external perturbations (on the microscopic or macroscopic scale). In terms of the first order functional derivatives, our approach gives a model- and material-independent, relativistic account of electromagnetic response functions that fully incorporates the effects of retardation, inhomogeneity, anisotropy and magnetoelectric coupling. Our core findings are:

1. A generalized expression for the Green function of the electromagnetic four-potential, Eq. (3.41).
2. The connection between classical electrodynamics of media and the Schwinger–Dyson or Hedin equations (Section 5.2).
3. The explicit formulae (5.54)–(5.57), by which one can calculate the 36 components of the field strength response tensor in terms of only 9 independent components of the fundamental response tensor.
4. The identification of physical response functions with *total* functional derivatives (Sections 4.2 and 6.1).
5. The universal relations between the physical response functions, cf. Eqs. (6.41)–(6.44) together with (6.45)–(6.48).
6. The existence of different but equivalent field expansions in terms of partial or total functional derivatives (Section 6.6).
7. The rederivation of well-known response relations as limiting cases of the universal response relations (Section 7).

The Functional Approach to electrodynamics of materials is exclusively based on the Maxwell equations and the interpretation of induced fields as functionals of the external perturbations. From this, all our conclusions and formulae follow analytically. As the Functional Approach is free of any assumption about the medium, it is particularly useful for studying conceptual issues,

such as general conditions for magnetoelectric coupling. On the other hand, the Functional Approach is computationally based on the explicit expression of all linear electromagnetic material properties in terms of the fundamental response tensor. In microscopic calculations, this fundamental response tensor is directly accessible from the four-point Green function through the Kubo formalism [9, 10]. Since typical first-principles methods such as the Hartree-Fock or the GW approximation can be grouped into a hierarchy of approximations for this four-point Green function [104], the Functional Approach is ideally suited for the ab initio calculation of electromagnetic material properties.

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